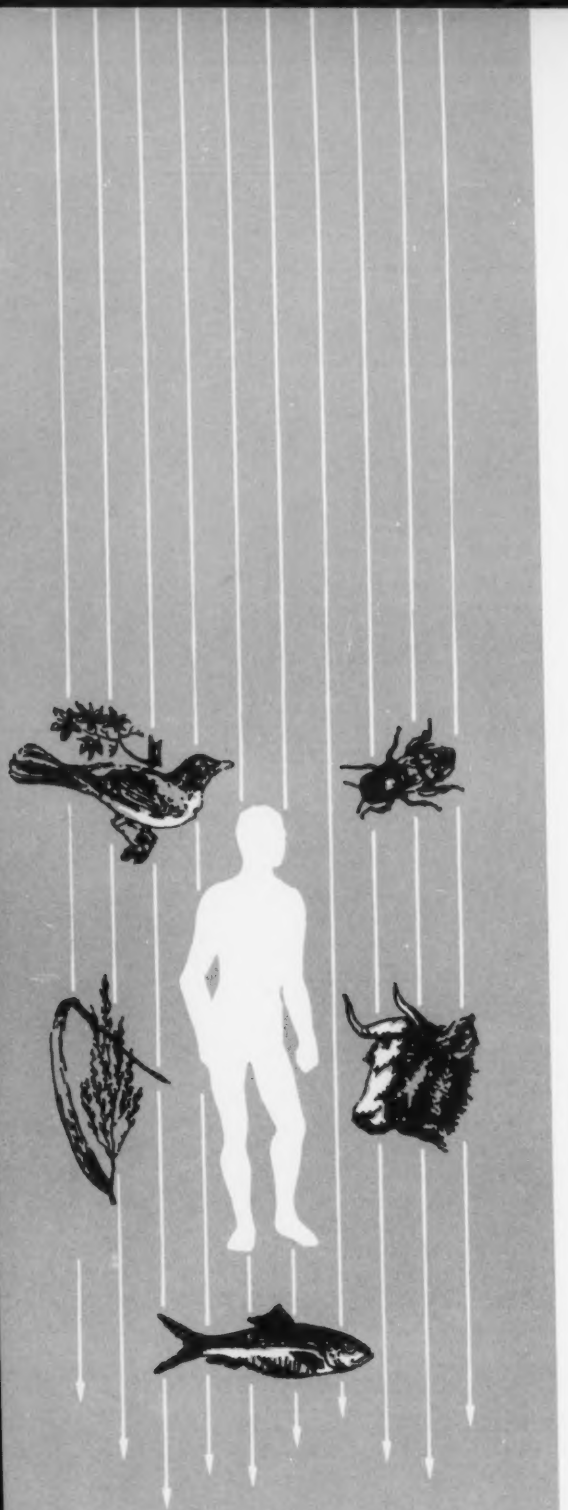

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The Monitoring Panel consists of representatives of the Agricultural Research Service, Animal and Plant Health Inspection Service, Extension Service, Forest Service, Department of Defense, Fish and Wildlife Service, Geological Survey, Food and Drug Administration, Environmental Protection Agency, National Marine Fisheries Service, National Science Foundation, and Tennessee Valley Authority.

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CONTENTS

Volume 11 March 1978 Number 4

Page

RESIDUES IN FOOD AND FEED

Organochlorine insecticide residues in bovine milk and manufactured milk products in Illinois, 1971–76 161
John L. Wedberg, Stevenson Moore III, Francis J. Amore, and Harold McAvoy

RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Variations in DDT concentration in muscle tissue of channel catfish, Ictalurus punctatus, *from the Des Moines River, 1971* 165
Ross Vivian Bulkley

p,p'-*DDE, polychlorinated biphenyls, and endrin in oldsquaws in North America, 1969–73* 170
Steven R. Peterson and Robert S. Ellarson

Mercury, cadmium, lead, and arsenic in sediments, plankton, and clams from Lake Washington and Sardis Reservoir, Mississippi, October 1975–May 1976 182
Richard E. Price and Luther A. Knight, Jr.

RESIDUES IN WATER

Residues of polychlorinated biphenyls, DDT, and DDT metabolites in Pennsylvania streams, community water supplies, and reservoirs, 1974–76 190
David A. Kurtz

GENERAL

DDT residues in rainwater in New Brunswick and estimate of aerial transport of DDT into the Gulf of St. Lawrence, 1967–68 199
Peter A. Pearce, Lincoln M. Reynolds, and David B. Peakall

APPENDIX 205

ACKNOWLEDGMENT 206

ANNUAL INDEX (Volume 11, June 1977-March 1978)

Preface 207
Subject Index 208
Author Index 216

Information for Contributors 218

RESIDUES IN FOOD AND FEED

Organochlorine Insecticide Residues in Bovine Milk and Manufactured Milk Products in Illinois, 1971-76

John L. Wedberg,[1] Stevenson Moore III,[1] Francis J. Amore,[2] and Harold McAvoy[3]

ABSTRACT

Monitoring activities were initiated in 1971 to survey the occurrence and levels of organochlorine insecticide residues in bovine milk and manufactured milk products in Illinois. Dieldrin residues were the most prevalent, and were found in 96 percent of the samples. Dieldrin also accounted for the highest average residue concentration (0.09 ppm). Only 0.3 percent of the samples contained illegal insecticide residues. Levels of DDT and lindane were generally declining, but those for dieldrin and heptachlor epoxide tended to remain constant.

Introduction

Use of chlorinated hydrocarbon insecticides on corn soil reached a peak in Illinois during 1967 *(2)* when an estimated 5,601,572 acres of farmland were treated (Table 1).

TABLE 1. *Number of corn acres in Illinois treated with different types of soil insecticides, 1964 through 1976*

Year	Chlorinated Hydrocarbons	Organophosphates and Carbamates
1964	4,009,303	81,822
1965	4,544,432	189,352
1966	5,116,605	326,592
1967	5,601,572	602,721
1968	5,170,726	1,091,143
1969	4,517,931	1,990,138
1970	3,844,740	2,763,547
1971	2,723,119	3,418,920
1972	1,933,089	3,852,239
1973	1,737,510	3,960,543
1974	1,886,042	4,128,300
1975	916,480	4,586,390
1976	935,550	5,428,870

Use of these insecticides has steadily declined since that time because of the development of insect resistance and of farmers' concern about persistence and long-term effects of aldrin, heptachlor, and chlordane residues. The chlorinated hydrocarbons generally have been replaced by organophosphates and carbamates. A study by Moore et al. *(1)* indicated that dieldrin residues were most likely to exceed the action level of the Food and Drug Administration, U.S. Department of Health, Education, and Welfare, on dairy farms which had recently been treated with aldrin. The study showed that hay and oat straw bedding supplied significant amounts of dieldrin to dairy cattle even when this material had been grown on farms with no history of aldrin treatments. Air movement of soil particles containing dieldrin was the reason given for roughage contamination on these farms.

A few Illinois dairy farms have produced milk containing illegal dieldrin residues within recent years *(1)*. Occasionally, this resulted from accidental contamination of feed, but in the majority of cases, the insecticide had been used in accord with the label.

The University of Illinois Cooperative Extension Service and Illinois Natural History Survey (Illinois State Department of Registration and Education) advised against the use of DDT on dairy farms in 1951 because of potential residues in milk. The use of aldrin, chlordane, dieldrin, endrin, heptachlor, or lindane has not been recommended for dairy farms since 1965 or for other farms since 1970. These actions preceded use cancellations by the U.S. Environmental Protection Agency. In September 1971, the Illinois State Department of Public Health announced that it was no longer legal for dairy farmers to store or use these insecticides for agricultural purposes on their farms. The Illinois State Department of Public Health began monitoring milk supplies for insecticide residues in 1971. This report summarizes these findings. All residues expressed in this paper are reported on a fat basis.

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[2] Illinois Water Survey, formerly, Illinois Department of Public Health, Division of Laboratories, Box 232, Urbana, IL.
[3] Illinois Department of Public Health, Office of Consumer Health Protection, Division of Milk Control, 535 West Jefferson St., Springfield, IL 62761.

Sampling Procedure

Samples were drawn at random from tank trucks of milk or from manufactured milk products in the plant and were analyzed for insecticide residues at either the Springfield or Chicago laboratories of the Illinois Department of Public Health. Each sample of raw milk represents at least eight farms; a manufactured milk product sample could include milk from a few to several hundred farms.

Method

EXTRACTION

Butterfat for residue analysis was extracted by placing 100 ml of milk, 30 g of grated cheese, or 50 g of melted ice cream in a 1-liter separatory funnel and adding 50 ml of 20 percent potassium carbonate, 100 ml of ethanol, and 50 ml of diethyl ether. This mixture was shaken briefly and 80 ml of Nanograde hexane was added. The resulting mixture was shaken approximately 200 times and allowed to stand until the phases separated. The lower aqueous phase was drained and discarded. For cheese samples both phases were transferred to another separatory funnel before separation of layers to remove the liquid from cheese solids. The lower aqueous phase was drained and discarded. The ether-hexane (organic) phase was washed three times with 100 ml of water to remove ethanol. Water was poured through the organic phase without shaking and the wash was discarded. Any emulsion formed was broken by adding about 5 ml of ethanol. The ether-hexane phase was transferred to a 250-ml beaker and placed on a sand bath maintained at 58°C in a fume hood; the volume was reduced to approximately 10 ml. Approximately 10 g of anhydrous granular sodium sulfate was added to remove water, the solution was transferred to a 100-ml beaker, and the remaining solvent was removed by passing a gentle stream of air over the solution.

ACETONITRILE PARTITIONING

A 1-g portion of butterfat was weighed into a 25-ml Erlenmeyer flask. The fat was dissolved in 15 ml of Nanograde hexane and transferred with rinsing to a 250-ml separatory funnel. The hexane was extracted four times with 30-ml portions of Nanograde acetonitrile saturated with hexane. The acetonitrile (lower layer) was drained into a 1-liter separatory funnel; after the fourth acetonitrile extraction, the hexane was discarded. The 120 ml of acetonitrile was transferred to a 250-ml separatory funnel and shaken with 10 ml of Nanograde hexane. After the lower acetonitrile layer was returned to the 1-liter separatory funnel, 500 ml of distilled water and 100 ml of saturated sodium chloride solution were added to salt out pesticides. The acetonitrile-water was extracted with 100 ml of 10 percent diethyl ether in hexane. The lower acetonitrile-water phase was discarded after separation. The ether-hexane phase was washed three times by passing 100-ml portions of distilled water through the ether-hexane to wash out traces of acetonitrile. Water washes were discarded. The ether-hexane was quantitatively transferred to a 250-ml beaker, with the aid of a hexane rinse, and placed on a sand bath to evaporate to about 10 ml. Approximately 5 g of sodium sulfate was added to remove traces of water.

COLUMN CHROMATOGRAPHY

Pesticides were separated and further cleaned by using glass columns, 250 × 20 or 330 × 22 mm, containing 80–100-mesh Florisil. The Florisil was heated at 450°C for 8 hours and stored at 130°C before use. Thirty g of Florisil was placed in the column and 0.5 inch of anhydrous sodium sulfate was added. The column was pre-eluted with 50 ml of acetone, 25 ml of anhydrous diethyl ether, 25 ml of water-saturated diethyl ether, and 25 ml of petroleum ether.

The pesticide extract was transferred to the Florisil column with three 1-ml portions of hexane. α-BHC, lindane, heptachlor, aldrin, heptachlor epoxide, γ-chlordane, α-chlordane, *p,p'*-DDE, *p,p'*-TDE, and *p,p'*-DDT were eluted with 120 ml of 6 percent diethyl ether in petroleum ether. Dieldrin and endrin eluted with 120 ml of 15 percent diethyl ether in petroleum ether. Eluates were collected in 250-ml beakers and reduced to approximately 5 ml by air evaporation in a fume hood. Samples were dried with about 2 g of sodium sulfate, transferred with washings of the eluting solvent to a 10-ml volumetric flask, and diluted to volume.

GAS-LIQUID CHROMATOGRAPHY

Pesticides were determined by injecting 5-μl samples into a Bendix Model 2500 gas chromatograph equipped with a Ni-63 electron-capture detector and a 6-ft X ¼-inch id glass column containing 1.95 percent QF-1/1.25 percent OV-17 on 100–120-mesh Supelcoport. Instrument operating conditions were:

Injection port temperature	225°C
Column temperature	190°C
Transfer line temperature	225°C
Detector temperature	275°C
Nitrogen flow rate	30 ml/minute
Sensitivity so that 100 pg of dieldrin gave a 30 percent recorder response with a noise level of about 0.5 percent	

A standard solution containing 0.01 ng/μl each of α-BHC, lindane, heptachlor, aldrin, heptachlor epoxide, γ-chlordane, and α-chlordane and 0.02 ng/μl each of *p,p'*-DDE, dieldrin, endrin, *p,p'*-TDE, and *p,p'*-DDT was used for retention time comparison and quantitation. Quantitation was based on peak height comparison between a standard and a sample.

EFFICIENCY MONITORING

Recovery efficiency of the 12 pesticides listed above was

continually monitored by using a sample of previously extracted butterfat to which all 12 pesticides were added to give final concentrations equivalent to those in the quantitation standard. Whenever recovery efficiencies decreased below 80 percent, final results were corrected for percent recovery. Procedural adjustments were made whenever recoveries decreased to less than 60 percent. This usually consisted of Florisil column modification. Aldrin had consistently lower recoveries than did the other insecticides; this was expected from its unfavorable partition coefficient from hexane to acetonitrile. Column packing was changed and the detector was cleaned as needed, based on reduced resolution and sensitivity.

PCBs, if present at levels above 0.5 μg/g fat, could be detected without procedure modification. No PCBs were detected in any samples. If PCBs had been present, it would have been necessary to modify the procedure to separate the pesticides and PCBs and prevent errors in the calculated pesticide levels.

Results and Discussion

Detailed results of the milk monitoring program for insecticides in 1971-76 are given in Table 2. Dieldrin was the most prevalent insecticide: it was found in 96 percent of the samples. Other insecticides which occurred in many of the samples were heptachlor epoxide, 93 percent; lindane, 73 percent; chlordane, 69 percent; and DDT, 48 percent. No other insecticides were found in significant amounts (>0.01 ppm) in these samples. Dieldrin also accounted for the highest average residue concentration in milk: 0.09 ppm; followed by heptachlor epoxide, 0.04 ppm; chlordane, 0.03 ppm; DDT, 0.01 ppm; and lindane, 0.01 ppm. DDT is the only insecticide with an official tolerance (1.25 ppm fat basis, 0.05 ppm whole milk basis) established in milk. Dieldrin was found in quantities above the action level in one sample during 1973, lindane exceeded the action level in one sample during 1974, and chlordane exceeded the action level in one sample during 1976. Therefore, in 6 years of sampling milk and manufactured milk products (1169 samples), only 0.3 percent of the samples contained illegal insecticide residues.

Too few samples were obtained in 1971 and 1972 for use in determining trends. However, the large number of samples obtained during 1973-76 makes it possible to discuss trends for insecticide levels in milk. Levels of DDT and lindane have generally declined since 1973, but dieldrin and heptachlor epoxide levels have not declined (Fig. 1). No organochlorine insecticide showed a definite upward trend in milk. During this study, combined chlorinated hydrocarbon residues in milk and manufactured milk products averaged 0.18 ppm (Table 2). The level of combined residues declined after 1973 (Fig. 1).

These data show that the occurrence of illegal residues of organochlorine insecticides in Illinois milk is rare. Levels are below the tolerance established by the Environmental Protection Agency or action levels established by the Food and Drug Administration. This trend is expected to continue as the use of insecticides such as aldrin, dieldrin, chlordane, and heptachlor for agricultural purposes is discontinued.

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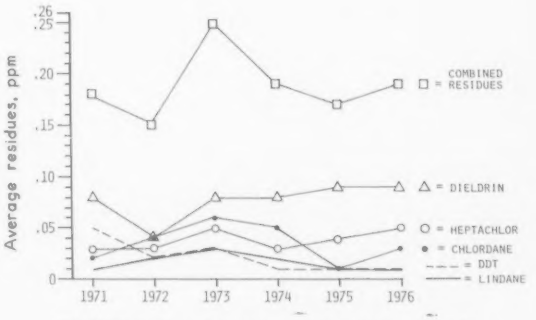

FIGURE 1. *Trends in insecticide residues in milk and in manufactured milk products, Illinois—1971-76*

TABLE 2. *Insecticide residues in samples of bovine milk and manufactured milk products, by year, Illinois—1971-76*

Insecticide[1]	No. Positive	Percent Positive	Avg. ppm	Percent Samples 0.01-0.10 ppm	Percent Samples 0.11-0.20 ppm	Percent Samples 0.21-0.30 ppm	Percent Samples 0.31+ 0.01–0.10 ppm
1971 (13 SAMPLES)							
Chlordane	5	39	0.02	100	0	0	0
DDT	12	92	0.05	92	8	0	0
Dieldrin	13	100	0.08	85	15	0	0
Heptachlor epoxide	13	100	0.03	100	0	0	0
Lindane	1	8	trace	100	0	0	0
Total	13	100	0.18	—	—	—	—
1972 (34 SAMPLES)							
Chlordane	34	100	0.04	100	0	0	0
DDT	31	91	0.02	97	3	0	0
Dieldrin	34	100	0.04	100	0	0	0
Heptachlor epoxide	30	88	0.03	100	0	0	0
Lindane	30	88	0.02	100	0	0	0
Total	34	100	0.15	—	—	—	—
1973 (153 SAMPLES)							
Chlordane	135	88	0.06	90	10	0	0
DDT	141	92	0.03	98	2	0	0
Dieldrin	141	92	0.08	65	31	3	1
Heptachlor epoxide	142	93	0.05	89	11	0	0
Lindane	130	85	0.03	96	2	2	0
Total	153	100	0.25	—	—	—	—
1974 (186 SAMPLES)							
Chlordane	143	76	0.05	84	16	0	0
DDT	122	64	0.01	98	2	0	0
Dieldrin	173	92	0.08	72	25	3	0
Heptachlor epoxide	162	86	0.03	98	2	0	0
Lindane	157	83	0.02	97	1	1	1
Total	186	98	0.19	—	—	—	—
1975 (283 SAMPLES)							
Chlordane	69	24	0.01	93	7	0	0
DDT	48	17	0.01	77	17	6	0
Dieldrin	271	96	0.09	72	24	4	0
Heptachlor epoxide	265	94	0.04	99	1	0	0
Lindane	259	92	0.01	100	0	0	0
Total	283	100	0.17	—	—	—	—
1976 (500 SAMPLES)							
Chlordane	421	84	0.03	98	1	0	1
DDT	213	43	0.01	98	2	0	0
Dieldrin	494	99	0.09	65	34	1	0
Heptachlor epoxide	481	96	0.05	97	3	0	0
Lindane	280	56	0.01	99	1	0	0
Total	500	100	0.19	—	—	—	—
SUMMARY, 1971-1976 (1,169 SAMPLES)							
Chlordane	807	69	0.03	94	6	0	0
DDT	566	48	0.01	96	4	0	0
Dieldrin	1126	96	0.09	69	29	2	0
Heptachlor epoxide	1093	93	0.04	97	3	0	0
Lindane	857	73	0.01	99	1	0	0
Total	1169	100	0.18	—	—	—	—

[1] α- and γ-chlordane residues are combined as chlordane; DDT, DDE, and TDE are combined as DDT; lindane and BHC are combined as lindane.

RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Variations in DDT Concentration in Muscle Tissue of Channel Catfish, Ictalurus punctatus, *from the Des Moines River, 1971*[1]

Ross Vivian Bulkley[2]

ABSTRACT

Concentrations of DDT in muscle tissue of channel catfish, Ictalurus punctatus, *from the Des Moines River, Iowa, were compared in relation to length, age, sex, and fat content. Residue concentrations were not correlated with sex. Although residue concentrations were significantly correlated with size-related factors and fat content, they varied widely. Therefore, predictions of DDT concentrations in individual catfish on the basis of length, age, or muscle fat content may be grossly inaccurate.*

Introduction

Accumulation of pesticides in fish at a given exposure is often envisioned as a simple function of age, body size, or fat content. Old, large, or fat fish are expected to contain higher residue concentrations than are young, small, or lean fish. The many exceptions and wide variations involved in the relation of pesticide concentrations to size-related factors in individual fish are frequently overlooked. Differences in pesticide concentrations sometimes exceed 100 percent among fish of similar size, age, or fat content *(3,7)*, and pesticide levels may decrease with increased age or fat content *(1, 6)*. Data on concentrations of DDT in muscle tissue of individual channel catfish, *Ictalurus punctatus*, obtained during a study of dieldrin contamination *(5)* provided an additional example of the variability in pesticide concentrations in relation to age, length, and fat content of the fish.

Methods

Channel catfish were collected from the Des Moines River near Fraser, Iowa, from April to October 1971, with baited hoop nets and electrofishing gear. The length of each fish was measured at capture. Fish were wrapped in aluminum foil and frozen until chemical analysis. Dorsal muscle tissue of all catfish collected in June was analyzed individually. On each collection date in other months, the fish in each of two length groups, 200-299 mm and 300-399 mm, were pooled for analyses. Percentage fat content of muscle tissue was determined by the rapid modified Babcock method *(2)* on individual samples. Sex was determined by examination of gonads, and age was determined from pectoral spine sections.

Muscle tissue samples were analyzed for DDT content according to the *Pesticide Analytical Manual (4)*. A sample of 10-40 g of tissue was ground with 350 ml of 35 percent distilled water-acetonitrile solution in a high-speed blender for 10 minutes. The sample was filtered through fluted number 40 Whatman paper. The filtrate (260 ml) was transferred to a 1-liter separatory funnel; 10 ml of petroleum ether was added, and the mixture was shaken for 2 minutes. Six hundred ml of distilled water and 10 ml of a saturated saline solution were added to the sample and mixed thoroughly by vigorous tumbling for 15 seconds. The layers were allowed to separate, and the aqueous layer was discarded. The solvent layers were gently washed with two 100-ml portions of water. The washings were dis-

[1] Journal Paper No. J-8616 of the Iowa Agriculture and Home Economics Experiment Station, Ames, IA. Project No. 1928. Financed by a grant from the U.S. Department of Interior, Office of Water Resources Research (Agreements 14-31-0001-3515, -3815, -4015) under Public Law 88-379 and made available through the Iowa State Water Resources Research Institute to the Iowa Cooperative Fishery Research Unit. The Unit is jointly sponsored by the Iowa State Conservation Commission, Iowa State University of Science and Technology, and the Fish and Wildlife Service, U.S. Department of Interior.

[2] Iowa Cooperative Fishery Research Unit, Iowa State University, Ames, IA 50011.

carded, and the solvent layer was filtered through a 50-mm column of anhydrous sodium sulfate into a 100-ml graduated cylinder. The volume was recorded, and the sample was filtered through a 115-mm column of Florisil topped by a 25-mm column of anhydrous sodium sulfate. DDT was eluted from the column with 200 ml of a 6 percent mixture of diethyl ether in petroleum ether. The solvent was evaporated to 10 ml for injection into the chromatograph.

A Beckman Model GC-5 gas chromatograph equipped with a discharge electron-capture detector was used to separate and quantitate the compounds of interest. A 5 percent OV-210 column at 180°C and 1.5 percent OV-17/QF-1 column at 200°C were used. Helium flow was about 100 mm/min and attenuation was $2x10^3$. A column of different polarity, 4 percent SE-30/6 percent QF-1, with a gas flow of 120-mm/min, a temperature of 200°C, and attenuation of $2x10^3$ was used as a qualitative check. Identities of compounds were confirmed by comparing retention times on the two columns with that of a DDT standard. Recovery of DDT and its metabolites ranged from 85 to 95 percent. Values were not corrected for percent recovery. Concentrations were expressed in wet weight.

All data were transformed to a $\log_{10}$ base for calculating regression and correlation coefficients of DDT in relation to fat content, length, and weight.

Results

ANALYSES OF INDIVIDUAL FISH

In June, 32 fish (14 males and 18 females, 155 to 602 mm long) were collected just before the spawning season. Differences in concentration of total DDT (DDT, TDE, and DDE) in males and females were not significant. Males averaged 355 mm long and contained an average of 1221 μg/kg of ΣDDT (78-6336 μg/kg) in muscle tissue; females averaged 325 mm long and contained an average of 799 μg/kg (154-4117 μg/kg). The regression lines expressing $\log_{10}$ DDT versus $\log_{10}$ body length were not significantly different ($P=0.05$) between males and females in terms of slope b ($t=0.25$, $df=30$) or intercept a ($t=0.28$, $df=30$). For all subsequent comparisons, therefore, data on male and female fish were combined.

Mean concentrations of DDT, DDE, TDE, and ΣDDT increased with length of fish in the June sample, with minor exceptions (Table 1). All DDT values given hereafter refer to ΣDDT. The correlation between length and DDT concentration ($r=0.63$) was significant at the 0.01 level of probability. The absolute and relative variation in DDT concentration, however, increased with size of the mean. The standard deviation ranged from 26 percent of the mean (coefficient of variation) in the 100-199-mm length group to 73 percent in the 300-399 group. Although the mean DDT concentration in muscle was greater in large than in small fish, the range in concentrations was also greater. Confidence limits about the mean DDT concentration for the large fish included the mean of all small fish. For example, the 500-599-mm group contained an average of 3389 μg/kg ΣDDT, with a standard deviation of 2317. The 95-percent confidence interval was -296 to 7974 μg/kg. The mean of all other length groups fell within this interval. Concentrations of DDT in the four fish in the 500-599-mm length group ranged from 1285 to 6336 μg/kg, and both values were obtained from fish of equal length (515 mm). Within the six length groups, the DDT concentration in individual fish was 0.6 to 6.4 times greater than the concentration in other fish the same length or shorter. The geometric mean DDT concentration more closely indicated the central tendency of the data for each length group than did the arithmetic mean (Table 1).

Inasmuch as one source of variation in pesticide extraction and quantitation is the person conducting the analysis, the June sample was divided into two groups for analysis by two chemists (Fig. 1). Although the author would have preferred having the chemists analyze duplicate samples of the same fish, this was precluded by the expense of analysis and the need for a sizeable series of samples. Analyses conducted by chemist A provided a relation between DDT concentration and body length, using $\log_{10}$ transformed data, expressed by the equation: $\log_{10}$ DDT (μg/kg) = $-1.50292 + 0.34265 \log_{10}$ total length (mm). Chemist B obtained a relation expressed by the equation: $\log_{10}$ DDT (μg/kg) = $-2.06662 + 0.17648 \log_{10}$ total length (mm). Data from both analysts fit a single regression line ($P<0.05$) on the basis of slope b ($t=0.18$, $df=30$) and intercept a ($t=0.30$, $df=30$). Correlation coefficients for length versus ΣDDT, using $\log_{10}$ transformed data, were 0.74 for chemist A and 0.58 for chemist B; the average value for all 32 fish was 0.63. All other samples in the study were analyzed by chemist A.

Fish in the June sample ranged from 2 to 12 years (Table 2, Fig. 1). Mean concentration of DDT did not increase with age as uniformly as with body length, although the correlation coefficient was close to that for length ($r=0.65$) and was also significant at the 0.01 probability level. The range in DDT levels was greatest (1288-6336 μg/kg) in fish of 7 years. The coefficient of variation expressed as a percentage of the mean was relatively low for ages 2 and 3 (13 and 24 percent, respectively), and values increased up to age 7 (95 percent), the oldest age group represented by more than one fish.

Fat content is frequently considered a major determinant in how much pesticide residue a fish will contain because of the lipophilic properties of many pesticides. Muscle fat ranged from less than 0.1 to 11.9 percent in the 32 fish. Fat content was correlated ($P=0.01$) with both length ($r=0.73$) and age ($r=0.63$). Fat content was highest in fish of 6 and 7 years and 450-525 mm long. Fish in this group were in prime spawning condition, which probably ex-

TABLE 1. *Residues of DDT, TDE, DDE, and ΣDDT in channel catfish from Des Moines River, June 1971*

Length Group, mm	No. Fish	Residues, μg/kg				ΣDDT			
		DDT	TDE	DDE	ΣDDT	C.V., Percent[1]	Confidence Limit[2]		G.M.[3]
100-199	6	119 (28)	70 (30)	170 (44)	363 (96)	26	253	to 473	348
200-299	9	126 (89)	62 (28)	143 (87)	332 (151)	45	217	- 447	290
300-399	6	166 (131)	137 (89)	300 (234)	604 (443)	73	139	1069	459
400-499	6	297 (236)	256 (190)	447 (255)	1001 (659)	66	310	1692	772
500-599	4	1211 (926)	695 (673)	1483 (578)	3389 (2317)	68	-296	7074	2794
600-699	1	1182	994	1675	3851	—	-	-	-
	Mean:	333	222	450	1006	56	-	-	-

Note: Standard deviations are shown in parentheses.
[1] Coefficient of variation = 100 (standard deviation)/mean.
[2] 95-percent confidence limits.
[3] Geometric mean.

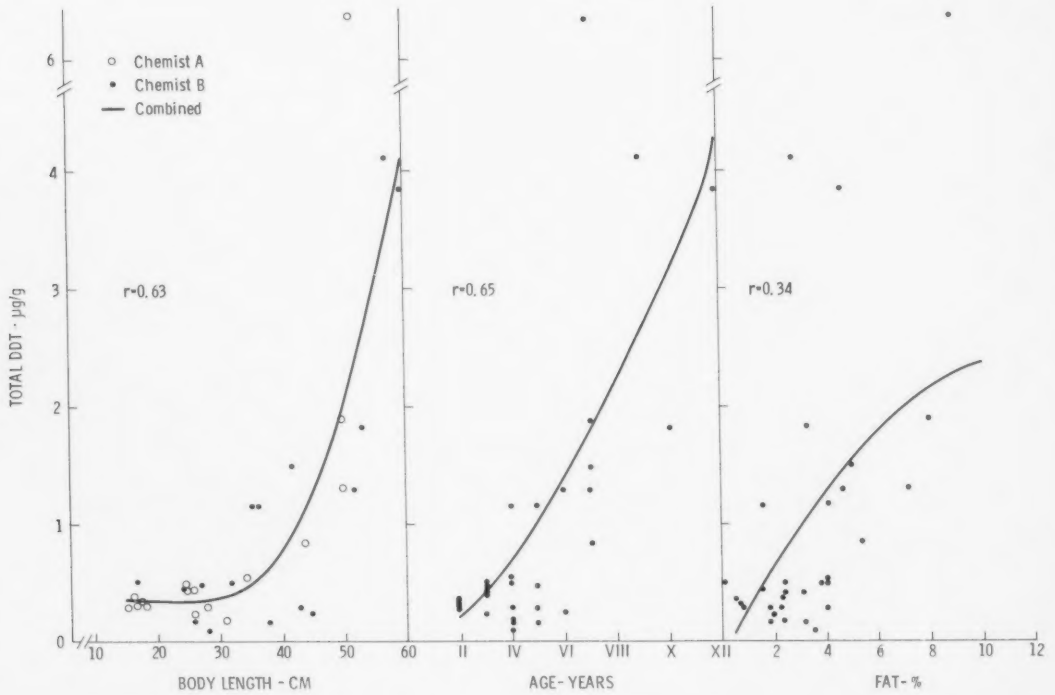

FIGURE 1. *Relation of ΣDDT to body length, age, and muscle fat content of channel catfish collected from Des Moines River, June 1971.*
(Lines on each graph express a second degree polynomial regression for the data.)

plained the high fat content in muscle tissue. Fat buildup in conjunction with gonad development is common in some species (*8*). Fish larger and older than 6 and 7 years contained less fat, and fish of 2 years and shorter than 200 mm contained the least. Correlation of muscle fat with DDT levels (r=0.34) was much lower than DDT versus length or age (Table 2) and the correlation value obtained was not significant at the 0.05 level of probability. To illustrate the low correlation, the fish with the highest fat content (11.9 percent) contained less DDT in the muscle tissue (261 μg/kg) than did four fish each containing less than 1 percent muscle fat. The range in DDT concentrations within each fat group (Table 2) was also large; in four groups the standard deviation exceeded the mean. Thus percentage fat content was of little value in predicting the amount of DDT present in muscle tissue of catfish of different lengths and ages.

TABLE 2. *ΣDDT in channel catfish of different ages and muscle fat content collected in June 1971*

Group	No. Fish	Mean[1]	S.D.	Confidence Limits[2]
		Residues, μg/kg		
Age, yr				
2	5	324 (322)	42	271 to 377
2	6	420 (407)	101	315 525
4	7	406 (291)	363	71 741
5	4	511 (388)	444	-195 1217
6	2	733 (579)	724	-5732 7278
7	5	2361 (1799)	2253	-434 5156
9	1	4117 -	-	- -
10	1	1819 -	-	- -
12	1	3851 -	-	- -
Fat, percent				
<0.1-0.9	4	364 (355)	104	199 529
1.0-1.9	5	411 (351)	436	-130 952
2.0-2.9	6	978 (505)	1541	-639 2595
3.0-3.9	5	590 (338)	708	-287 1467
4.0-4.9	6	1260 (854)	1331	-136 2656
5.0-5.9	2	1156 (1112)	450	-2884 5196
7.0-7.9	1	1288	-	- -
8.0-8.9	1	1871	-	- -
9.0-9.9	1	6336	-	- -
11.0-11.9	1	261	-	- -

[1] Geometric means in parentheses.
[2] 95-percent confidence limits.

TABLE 3. *Correlation of ΣDDT with length, age, and muscle fat content in channel catfish, 1971*

Variable	r Value	R Value
JUNE (ALL LENGTHS)		
Length	0.63	
Age	0.65	
Fat	0.34[1]	
Length, age		0.65
Length, fat		0.66
Age, fat		0.66
Length, age, fat		0.67
APRIL-OCTOBER (300-399 mm)		
Length	0.30[1]	
Age	0.34[2]	
Fat	0.31[1]	
Length, age		0.36[2]
Length, fat		0.37[2]
Age, fat		0.44
Length, age, fat		0.44

NOTE: Values are significant at the 0.01 level of probability unless otherwise noted.
[1] Nonsignificant at 0.05 level of probability.
[2] Significant at 0.05 level of probability.

Comparison of two or more size-related factors with DDT concentration accounted for little additional variation in the relationships (Table 3). Length, age, and fat accounted for 40, 42, and 11 percent, respectively, of the variation in DDT levels, but the amounts were not additive. When two, or all three, factors were considered together, a maximum of only 45 percent (R=0.67) of the variation in DDT could be accounted for.

SEASONAL SAMPLES (APRIL-OCTOBER)

Samples collected from April to October provided additional observations on the relation of DDT to length. Pooled samples of fish 200-299 mm long contained the highest DDT concentrations in April and May and the lowest in August and September (Table 4). In fish 300-399 mm long analyzed individually, the DDT concentration was also greatest in April. When one specimen containing 3218 μg/kg DDT was omitted from the July samples of fish measuring 300-399 mm, the trend in DDT concentration in both size groups was similar, ranging from highest in April to lowest in August. No significant difference was evident between seasonal mean DDT levels in fish of the two length groups (t=0.253, df=12); however, as in the June sample, fish 300-399 mm long averaged slightly more DDT than did those measuring 200-299 mm long (569 versus 515 μg/kg).

Comparison of DDT levels in the two length groups within each month from April to October revealed only minor differences, with one exception. The 95-percent confidence limits for the mean of the 300-399-mm group included the mean DDT levels of the 200-299-mm group in all months except April. The mean concentration (1448 μg/kg) of DDT in the shorter length group in April was unusually high. Because tissues of the April sample of 15 fish were combined for DDT analysis, variation among individual fish within the sample could not be measured. Even though the mean monthly DDT concentration in muscle tissue of the 300-mm fish which were anlyzed individually ranged from 798 μg/kg in April to 179 μg/kg in August, monthly means for the April-October period were not significantly different (F=1.83; df=6, 33).

Discussion

Pesticide concentrations in fish frequently do not bear a simple and direct relation to body condition, size, age, or fat content. Kleinert et al. *(6)* reported that the relation of DDT levels to percentage fat was not significant in fish from most Wisconsin water samples. Anderson and Fenderson *(1)* found significantly greater concentrations of DDT in high-fat Atlantic salmon *(Salmo salar)* than in low-fat fish at ages 3+ and 4+ but not at age 5+. They recommended stratification of sampling by sex, age, and fat content for pesticide analysis. Bulkley et al. *(3)* found that dieldrin concentrations in muscle tissue of the same channel catfish used in this paper were significantly correlated with body length (r=0.46) and age (r=0.60) only if fish of all lengths and ages were included in the analyses. When only fish shorter than 400 mm and younger than age 6 were considered, dieldrin concentration was not related to length and age. Dieldrin was significantly correlated

TABLE 4. *Seasonal concentrations of ΣDDT (DDT, TDE, DDE) in two length groups of channel catfish from Des Moines River, April-October 1971*

Month	200-299 mm Long			300-399 mm Long			C.V., Percent[1]	Confidence Limits[2]		
	No. Fish	Mean	SD	No. Fish	Mean	SD				
April	15	1448	-	9	798	518	65	407	to	1289
May	7	884	-	8	689	769	112	46		1332
June	9	332	151	6	604	443	73	139		1069
July	6	231	-	7	780	1083	139	-221		1781
August	4	101	-	3	179	78	43	-16		374
September	3	224	-	4	696	303	43	212		1180
October	3	385	-	3	239	74	31	54		424

[1] Coefficient of variation = 100 (standard deviation)/mean.
[2] 95-percent confidence limits.

with muscle fat content ($r=0.68$, $P=0.01$) only if the longest and oldest catfish was removed from the sample. When all fish were included, the correlation coefficient was 0.27. The correlation of ΣDDT concentration with length, age, or fat content was slightly higher than the correlation between dieldrin and these variables in the same fish ($r=0.68$, 0.68, 0.45, respectively), but exceptions to these relationships were common. Other size-related variables, such as metabolic rate or individual variations in diet or in development of detoxifying enzyme systems, are undoubtedly involved in DDT and dieldrin accumulation in wild catfish. Hence predictions of concentrations of DDT or dieldrin in muscle of individual channel catfish on the basis of length, age, or muscle fat content may be grossly inaccurate, and generalities about any relationship should be made with caution.

Acknowledgment

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p,p'-DDE, Polychlorinated Biphenyls, and Endrin in Oldsquaws in North America, 1969-73

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ABSTRACT

Organochlorinated compounds were monitored in oldsquaws (Clangula hyemalis) *and their food from Lake Michigan between October and May, 1969-72; in adult oldsquaws, eggs, young, and food from northwest Hudson Bay in 1971; and in oldsquaws from five wintering areas other than Lake Michigan in 1971-73. Analyses were conducted on 300 carcasses, 14 wings, 29 gullet samples, and 11 clutches. Average residues in carcasses from Lake Michigan ranged from 4 to 107 ppm PCBs, 2 to 42 ppm DDE, and <0.1 to 0.7 ppm endrin. Differences in DDE levels occurred between several sex and age classes during December on Lake Michigan; these differences were not apparent in the spring. Increases in DDE and PCB residues for oldsquaws occurred on Lake Michigan between December and May. DDE residues in the wing and carcass were significantly correlated. Residues were relatively low in oldsquaw foods from Lake Michigan; concentration factors between the food and the ducks varied between 1× and 22×, depending on the date and compound. Organochlorinated residues were lower in Arctic than in Lake Michigan food samples. DDE in paired male and female oldsquaws was highly correlated, as was DDE in females and clutches. Eggshell thickness had declined 4.5 percent compared with eggs collected before 1947. Residues were highest in oldsquaws wintering on the Great Lakes and lowest in oldsquaws from coastal areas.*

Introduction

An exploratory study on Lake Michigan indicated that several trophic levels had been contaminated *(16)*. This paper reports residues of *p,p'*-DDE, endrin, and polychlorinated biphenyls (PCBs) in oldsquaw *(Clangula hyemalis)* adults, eggs, young, and food. Various organochlorinated compounds were measured in this study, but because residue identities could not be confirmed by mass spectrometry, and because PCB levels were relatively high, results are not included for TDE, DDT, dieldrin, BHC, and heptachlor epoxide. These data are available from the senior author on request.

Oldsquaws wintering on Lake Michigan are suitable for monitoring contamination on the lake. The species arrives on Lake Michigan in November and remains there until April or May *(7)*. Presumably, then, any detectable increase in pesticide body burdens after November would come from within the lake.

While on the lake the species feeds almost entirely on a bottom dwelling deep-water amphipod, *Pontoporeia affinis (7, 25)*. This invertebrate is also the predominant food of the commercially important lake whitefish *(Coregonus clupeaformis) (7, 20);* thus this food organism in common with both consumers allowed authors to measure concentration in a bird and to compare findings with other studies on a fish at the same trophic level.

Several age and sex classes are discernible in the oldsquaw *(1, 7, 24)*. This allowed authors to test for differences in body burdens between sex, age, and time.

Since the oldsquaw is an Arctic nester, the Lake Michigan population spends about half the year in a relatively polluted environment and the remainder in a relatively clean environment. Thus shifts in body burdens caused by changes in diet contamination were investigated, and the transfer of pesticides from a wintering to a breeding area was explored.

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Methods

SAMPLE COLLECTION

Samples collected on Lake Michigan were obtained from commercial fishing operators, who found the birds drowned in gill nets. All other birds were killed by shotgun except those collected on Lake Huron. The latter samples were found dead on a beach; an autopsy performed by the Rose Lake Laboratory of the Michigan Department of Natural Resources indicated that the birds had died of hemorrhagic enteritis of undetermined cause.

Samples were catalogued between October 30 and August 10, 1969-73. They were either examined and prepared immediately or frozen until dissection and preparation. Sex and age were confirmed during dissection. Hanson's *(10)* classification was adopted for age categories: juvenile oldsquaws are less than fully grown and less than 1 year old; subadults are more than 1 year old and essentially fully grown although the majority of the cohort has not completed its first breeding season; adults are at least 2 years old, fully grown, and most members of the cohort have completed one or more breeding seasons. In this paper, adults and subadults combined in one class are called matures.

Carcasses were prepared for chemical analysis according to the following procedure: the head was severed, wings were clipped at the distal end of the humerus, feet were removed at the tarso-metatarsal joint, and feathers were sheared with scissors as close to the skin as possible. After the gastrointestinal and reproductive tracts were removed, the carcass was wrapped in aluminum foil and frozen until chemical analysis.

Food samples were removed from the esophagi of oldsquaws. Only *Pontoporeia affinis*, with some adhering inorganic material, was analyzed from the Lake Michigan samples. Since little food was noted in oldsquaws collected in the Arctic, all food found in the esophagi was composited. Food samples were placed in glass jars previously rinsed with acetone, and sealed with aluminum foil, capped, and frozen until analysis.

Eggs from clutches incubated less than 1 week were wrapped in aluminum foil and frozen for shipment. In the laboratory, the eggs were slightly thawed, and the contents were placed in glass jars previously rinsed with acetone. The jars were then sealed with aluminum foil, capped, and frozen until analysis of the contents.

Wings were prepared for analysis by clipping the feathers as close to the skin as possible. The samples were then wrapped in aluminum foil and frozen for analysis.

EGGSHELL MEASUREMENTS

Weight, length, and breadth of all fresh eggs were measured before freezing. After the contents had been removed, the shells were washed and air-dried to a constant weight; then shell thickness was measured at the equator with a modified Starrett Model 1010M micrometer. Measurements on pre-1947 eggs were obtained from clutches deposited in the Field Museum of Natural History in Chicago and the Museum of Vertebrate Zoology, University of California, Berkeley, California.

CHEMICAL ANALYSIS

All samples were anlyzed at WARF Institute, Inc., Madison, Wisconsin, by a gas chromatographic procedure. The entire carcass, wing, egg contents of a composite clutch, a composite brood, or composite food sample was homogenized in a Hobart food chopper. A sample was removed, weighed, dried at 40°C for 72-96 hours, reweighed, ground with sodium sulfate, and extracted for 8 hours on a Soxhlet extractor using 70 ml of ethyl ether and 170 ml of petroleum ether. An aliquot of the sample was cleaned up on a column of previously standardized Florisil. Typical elutions were 150 ml of 5 percent ethyl ether in petroleum ether, followed by 240 ml of 15 percent ethyl ether in petroleum ether. Compounds were separated and detected on a Barber-Colman Pesticide Analyzer Model 5360 with two columns: one 4-ft x 4-mm column was packed with 5 percent DC-200 on 60–70-mesh Cromport XXX. The column temperature was 200°C, and the nitrogen flow was one which gave *p,p'*-DDT a retention time of 6-8 minutes. The other similar-size column was packed with 3 percent OV-17 on 100–120-mesh Gas-Chrom Q with a column temperature of 195°C and a nitrogen flow which gave lindane a retention time of 1 minute.

The following procedure was used to quantitate PCBs: the samples were injected, and peaks on the DC-200 column which had retention times of TDE and DDT were measured and quantitated. The extract was then subjected to alkaline hydrolysis and re-injected, and the PCBs were quantitated against Aroclor 1254, using the peaks between TDE and DDT and the peak at DDT. Endrin was separated from the PCBs by the initial Florisil chromatography.

Results

LAKE MICHIGAN

Relationship of Body Burden to Age/Sex Class and Time—Of the compounds measured, DDE and PCB residues were generally the highest; endrin residues were relatively low (Table 1). In December, mean DDE levels varied from 4.9 ppm in juvenile males and females to 13.8 ppm in mature males. Adult females averaged 7.0 ppm; this was significantly lower than the average residue level recorded from subadult females ($\bar{x}$ = 10.1 ppm; $P<0.05$) and mature males ($\bar{x}$ = 13.8 ppm; $P<0.01$).

Average December PCB levels generally were higher than December DDE levels but otherwise exhibited trends similar to DDE. Mature males contained the highest residues,

averaging 19 ppm; averages for juvenile males were the lowest, 5 ppm. Statistical tests similar to those performed on DDE showed similar levels of significance for PCBs between different age/sex classes.

No apparent trend was noted for endrin. Overall F-tests among age/sex classes were nonsignificant, and t-tests between selected groups did not yield any statistically significant information.

DDE and PCB residues were much higher ($P<0.05$) in March-April than in the previous December in all age/sex classes (Table 1). Significant differences between age/sex classes which had been apparent in early winter were not present in the spring.

In 1970-71, collections were expanded to monthly intervals in order to check yearly differences in residues, to confirm early winter differences in DDE and PCB residues between

TABLE 1. *Mean and standard deviation of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1969-70*

Sample Size	Age/Sex Class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
	1-15 DECEMBER 1969			
8	Mature male	13.8 ±5.3	19 ±14	0.2 ±0.1
8	Juvenile male	4.9 ±2.5	5 ±2	0.2 ±0.1
10	Adult female	7.0 ±3.6	9 ±4	0.1 ±0.1
4	Subadult female	10.1 ±0.8	14[1] ±4	0.2 ±0.1
7	Juvenile female	4.9 ±2.2	9 ±13	0.2 ±0.1
	22 MARCH - 22 APRIL			
13	Mature male	42.2 ±21.5	75 ±64	0.2 ±0.1
9	Juvenile male	34.7 ±13.4	69 ±38	0.4 ±0.3
12	Adult female	34.1 ±14.5	72 ±57	0.3 ±0.2
6	Subadult female	24.1 ±15.4	46 ±32	0.3 ±0.1
4	Juvenile female	18.5 ±5.3	30 ±14	0.2 ±0.1

[1] One value in the mean was 41; without this value, $\bar{x} = 4 \pm 2$.

TABLE 2. *Mean and standard deviation of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1970-71*

Sample Size	Age/sex class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
	DECEMBER			
4	Mature male	12.5 ±6.0	20 ±7	0.1 ±0.1
4	Juvenile male	2.9 ±1.0	4 ±1	0.9 ±1.1
4	Adult female	6.0 ±1.1	11 ±2	0.1 ±0.1
6	Subadult female	8.2 ±4.7	18 ±12	0.2 ±0.1
6	Juvenile female	2.1 ±0.9	8 ±10	0.2 ±0.1
	JANUARY			
2	Mature male	12.6 ±2.8	23 ±2	0.5 ±0.2
4	Juvenile male	5.0 ±2.8	14 ±17	0.4 ±0.2
4	Adult female	8.5 ±4.8	15 ±11	0.5 ±0.2
2	Subadult female	7.7 <0.1	14 ±4	0.6 ±0.1
4	Juvenile female	2.4 ±0.6	4 ±1	0.4 ±0.3
	FEBRUARY			
0	Mature male	-	-	-
4	Juvenile male	5.9 ±3.3	12 ±5	0.5 ±0.1
0	Adult female	-	-	-
0	Subadult female	-	-	-
4	Juvenile female	4.7 ±0.8	9 ±2	0.5 ±0.2

Sample Size	Age/sex class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
	MARCH			
4	Mature male	19.1 ±8.7	39 ±16	0.4 ±0.1
4	Juvenile male	10.1 ±2.7	25 ±14	0.4 ±0.1
4	Adult female	12.2 ±3.9	27 ±6	0.3 ±0.2
4	Subadult female	12.8 ±5.9	34 ±24	0.3 ±0.2
4	Juvenile female	7.8 ±3.8	25 ±22	0.2 ±0.1
	APRIL			
4	Mature male	14.8 ±10.1	89 ±55	0.2 ±0.2
4	Juvenile male	13.5 ±6.0	38 ±23	0.3 ±0.1
4	Adult female	15.0 ±7.8	52 ±12	0.8 ±0.7
5	Subadult female	25.4 ±7.9	71 ±41	0.3 ±0.1
4	Juvenile female	9.8 ±4.7	36 ±38	0.3 ±0.1
	MAY			
4	Mature male	30.9 ±11.6	86 ±69	0.2 ±0.1
6	Juvenile male	21.5 ±6.4	54 ±25	0.2 ±0.1
4	Adult female	32.1 ±18.0	85 ±40	0.3 ±0.1
1	Subadult female	30.9	115	0.4
4	Juvenile female	18.7 ±5.2	37 ±22	0.2 ±0.1

Note: All values were used in computing means; trace values were used as one-half the stated "less than" value and residues not detected were treated as zero.

age/sex classes, and to graph the apparent accumulation in DDE and PCB residues over the winter.

Early winter and spring levels for 1970-71 were similar to those recorded the previous winter (Table 2). For a particular age/sex class, average December DDE (except in juvenile females) and PCB levels were not significantly different between 1969 and 1970 ($P>0.05$). In March-April 1970-71, DDE and PCB levels were not significantly different ($P>0.05$) for any particular age/sex class.

The increases ($P<0.01$) of DDE and PCB residues in oldsquaws wintering on Lake Michigan between October and May (Fig. 1) are similar, but the increase is greater for PCBs. In birds starting the winter with relatively low residue levels (juveniles and adult females), the data fit the equation for a curved line ($Y = aX^b$) better than for a straight line ($Y = a + bX$). For DDE, juveniles had a linear coefficient of determination of 69 percent and a power coefficient of determination of 85 percent. For PCBs, these coefficients were 48 percent and 70 percent, respectively. Similarly, in adult females, these coefficients increased from 43 to 58 percent for DDE, and from 52 to 71 percent for PCBs. The data for subadult females and mature males did not suggest a better fit to a curved line equation because the coefficients decreased from 59 to 55 percent and from 29 to 26 percent in DDE for these respective age/sex classes.

These results suggest two relationships: that DDE and PCBs increase at a relatively constant rate throughout the winter; and that coefficients of determination are higher in younger birds than in older birds, which indicates that as a cohort ages, its members are exposed to varying amounts of contamination and consequently the variability in the data increases.

During January and February, it is often difficult to obtain sufficient samples of oldsquaws because ice makes it difficult to tend the nets. In 1972, the previous collection was augmented with some additional samples from these months (Table 3). These data largely confirm the results discussed in Table 2.

Comparison of Milwaukee Harbor and Lake Michigan Oldsquaws—A population of 5,000-10,000 oldsquaws apparently winters in Milwaukee Harbor *(29)*. A sewage treatment plant is located in the center of the harbor, and the bottom mud contains high levels of PCBs *(33)*. It was hypothesized that the oldsquaws feeding on the bottom organisms would reflect these high levels of PCBs.

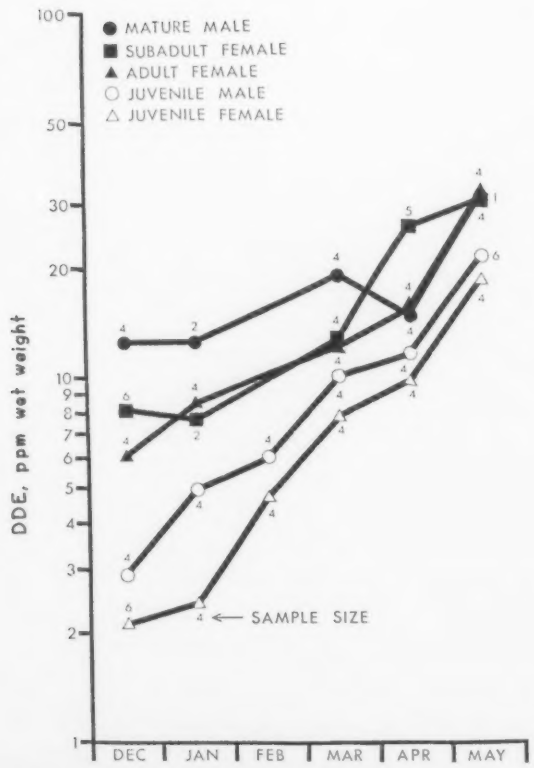

FIGURE 1. *Semilog plot of DDE residues in different age/sex classes of oldsquaws collected on Lake Michigan, 1970-71*

TABLE 3. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1972*

Sample Size	Age/sex class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
		JANUARY		
2	Mature male	19.2 13.0-26.4	50 30-70	0.4 0.2-0.5
2	Subadult female	9.7 6.3-13.0	18 11-25	0.7 0.4-1.0
		FEBRUARY		
2	Mature male	21.3 19.5-23.1	43 33-54	0.7 0.6-0.8
2	Adult female	8.2 5.3-11.1	18 13-23	0.6 0.5-0.8

TABLE 4. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected in Milwaukee Harbor, January 23, 1972*

Sample Size	Age/sex class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
4	Mature male	12.2 4.5-27.3	97 60-122	<0.1 ND-0.1
2	Juvenile male	9.5 9.2-9.8	82 80-84	0.3 0.1-0.5
4	Adult female	6.9 3.1-11.5	89 78-130	<0.1 ND-<0.1
2	Subadult female	14.3 13.0-15.6	107 69-145	0.2 ND-0.3

Note: ND = not detected = no peak observed. All values were used in computing means; trace values were used as one-half the stated "less than" value and ND was used as zero.

Twelve birds were collected in January 1972 (Table 4); PCB residues averaged 5-8 times higher in these oldsquaws than in oldsquaws obtained from Lake Michigan during January 1971, and 2-6 times higher than in oldsquaws collected in January 1972. DDE residues were about the same as in Lake Michigan, and mature males and subadult females contained higher concentrations than juvenile males and adult females, as in the Lake Michigan data. Endrin occurred in relatively low concentrations in the Milwaukee Harbor samples, as in Lake Michigan samples.

Correlation of DDE in Wings and Carcasses—Several investigators have determined a significant relationship between organochlorinated compounds or mercury in wings and various tissues of birds (*5, 11, 34*). Authors hypothesized that if a smiliar relationship could be established in oldsquaws between the wing and carcass, average monthly residues could be more precisely measured by pooling large numbers of wings of a given age/sex class for a particular month. In addition, the number of determinations could be reduced if authors assumed that sufficient variability was incorporated into the sample.

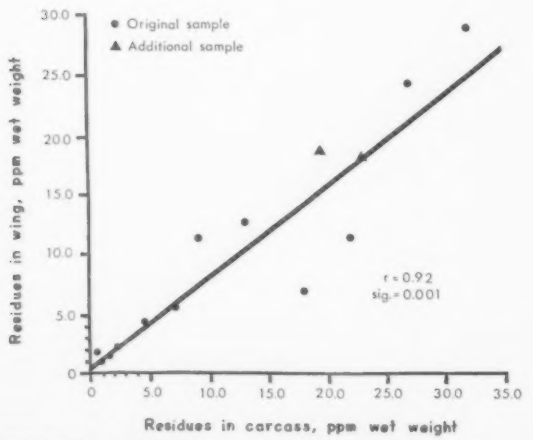

FIGURE 2. *DDE residues in wings and carcasses of mature male oldsquaws collected on Lake Michigan, 1970-71*

Figure 2 illustrates the positive correlation ($P<0.001$) between DDE in oldsquaw wings and carcasses. Residues in the carcasses selected ranged from 0.4 to 32.0 ppm. The relatively high correlation coefficient ($r = 0.91$) indicates that oldsquaw wings could be used to monitor pesticide burdens in the carcasses.

Residues in Food—Table 5 presents monthly averages of DDE, PCB, and endrin residues in amphipods taken from oldsquaw gullets. The data suggest that residues are relatively constant between December and May when old-

TABLE 5. *Mean and range of organochlorine pesticide and PCB residues in amphipod samples taken from the gullets of oldsquaws collected on Lake Michigan*

Month	Year	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
December	1969	0.62 0.56-0.68	1.26 1.18-1.37	0.04 0.04-0.04
December	1970	1.74 1.59-1.91	3.36 3.18-3.45	0.08 0.05-0.10
January	1971	0.87 0.60-1.23	1.66 1.15-2.38	0.06 0.05-0.08
February	1971	2.66 1.95-4.01	5.60 4.91-6.44	0.17 0.04-0.33
March	1970	1.21 1.04-1.50	2.84 2.02-3.69	0.08 0.07-0.10
March	1971	1.07 1.04-1.13	2.29 2.02-2.49	0.08 0.05-0.11
April	1971	0.81 0.12-1.28	2.57 2.23-2.82	0.06 0.05-0.08
May	1971	1.22 1.07-1.51	3.23 2.94-3.51	0.08 0.06-0.11
Overall average and range	1969-1971	1.27 0.12-4.01	2.85 1.15-6.44	0.08 0.04-0.33

NOTE: $n = 3$ for each monthly sample.

TABLE 6. *Biological magnification of organochlorine pesticide and PCB residues between amphipods and oldsquaws collected on Lake Michigan, 1970-71*

Month	Sample Size[1]	*p,p'*-DDE	PCBs	Endrin
December	A = 3 OS = 24	4	4	3
January	A = 3 OS = 16	8	8	8
February	A = 3 OS = 8	2	2	3
March	A = 3 OS = 20	12	13	4
April	A = 3 OS = 21	20	22	6
May	A = 3 OS = 19	21	21	3

[1]A = amphipods; OS = oldsquaws.

squaws were collected for analyses. The apparent buildup of DDE and PCBs in oldsquaws throughout the winter is not reflected in the food sample data. Where there is replicate sampling for a certain month in successive years, the concentration of a particular toxicant varies from 0 to 280 percent.

Biological magnification factors between oldsquaws and their food were computed for each compound by dividing monthly averages of the residues in oldsquaws with corresponding averages for the food (Table 6). Since samples of each age/sex class of oldsquaws were not constant within and between months, the magnification factors should be considered rough approximations.

Several interesting trends appear in these data. Biological magnification from amphipods to oldsquaws is not constant but can vary from 1× to 22×, depending on the compound or the time of year. Magnification factors for DDE and PCBs are similar and appear to increase through the winter and spring from 4× in December to 21× in May, but residues of these compounds remained relatively constant in the food during the same period. The magnification factor computed for endrin did not vary so widely over time. Thus these data suggest that biological magnification factors computed for PCBs and DDE are not static, but dynamic, and it may be more appropriate to determine rates of increase as a function of time for specific species, compounds, and localities.

NORTHWEST HUDSON BAY

Ducks were collected during the breeding season at Rankin Inlet and Eskimo Point, Northwest Terriories (NWT) in 1972 to help interpret difference in DDE and PCB residues in December oldsquaws on the wintering grounds; to measure any loss in body-burden residues that may occur on the breeding grounds; and to examine the passage of pesticides from the female to the young. Even though comparisons were made between levels observed on the breeding ground and those recorded on Lake Michigan, it should not be construed that the same populations were sampled. Since oldsquaws winter in several areas and breed throughout the Arctic, breeding populations may be composed of birds from one or several wintering areas.

Breeding Pairs—Breeding pairs were collected on tundra ponds as soon as the ice thawed around the edges and birds moved inland from the sea. Average residues for each compound were similar in paired males and females (Table 7). PCBs averaged 25 ppm in males and 18 ppm in females; DDE averaged 6.4 ppm in males and 6.5 ppm in females. Endrin averaged less than 0.1 ppm in both paired males and females.

Figure 3 indicates a strong relationship ($r = 0.86$; $P<0.01$) betweeen the levels of DDE in the male and female of a given pair. Alison *(1)* showed that oldsquaws pair early on the wintering grounds. Pesticide data from the Arctic reported here indicate that pair bonds maintained throughout the winter and migratory periods expose members of a pair to similar levels of contamination. Lake Michigan data collected just before spring migration also indicate similar residue levels in mature males and females (Fig. 1).

DDE residues in breeding pairs suggest that most of those birds obtained in the Arctic probably did not come from Lake Michigan. Of the 20 birds collected on Lake Michigan in May 1971, the lowest level of DDE recorded was

TABLE 7. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected at Eskimo Point, Diana River, and Rankin Inlet, Northwest Territory, 1971*

Class	Sample Size	Date	Residues, ppm Wet Weight		
			p,p'-DDE	PCBs	Endrin
Paired males	10	7-10 June	6.4 0.7-21.9	25 3-81	<0.1 ND-0.1
Paired females	10	7-10 June	6.5 0.6-19.8	18 3-44	<0.1 Tr-0.2
Females with clutches	11	29 June-11 July	4.7 0.1-16.0	24 1-95	<0.1 Tr-0.1
Composite clutches[1]	11	29 June-11 July	7.6 0.2-19.1	48 <1-172	0.1 ND-0.2
Females with broods	3	25 July-2 August	2.8 0.3-7.6	14 2-32	ND -
Composite broods[2]	3	25 July-2 August	2.1 0.2-3.1	25 1-63	<0.1 ND-<0.1
Molting females without broods	5	8 August	2.6 0.3-7.7	21 1-57	ND -
Subadult males[3]	4	10 July-8 August	2.9 0.6-6.8	15 <1-43	<0.1 ND-<0.1

Note: ND = not detected = no peak observed; - = range not calculated because no residue detected; all values were used in computing means; trace values were used as one-half the stated "less than" value and ND was used as zero.

[1] Clutches contained 1-8 eggs.

[2] Broods contained 3-5 young.

[3] These males still retained their juvenile plumage from the preceding year. After the summer molt, they could not be distinguished from males 2 years or older.

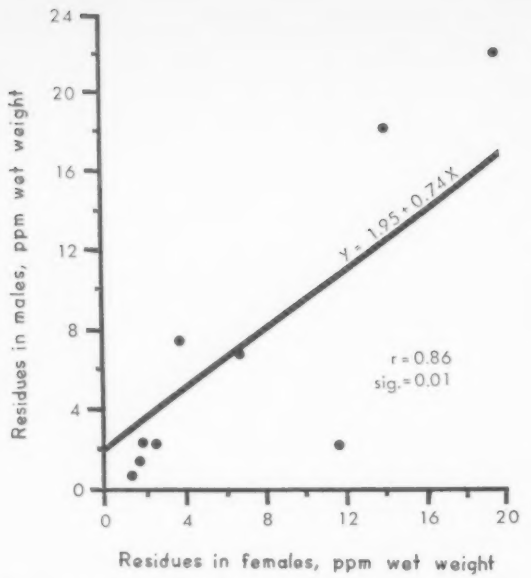

FIGURE 3. *DDE residues in pairs of oldsquaws collected at Rankin Inlet, Northwest Territory, June 1971*

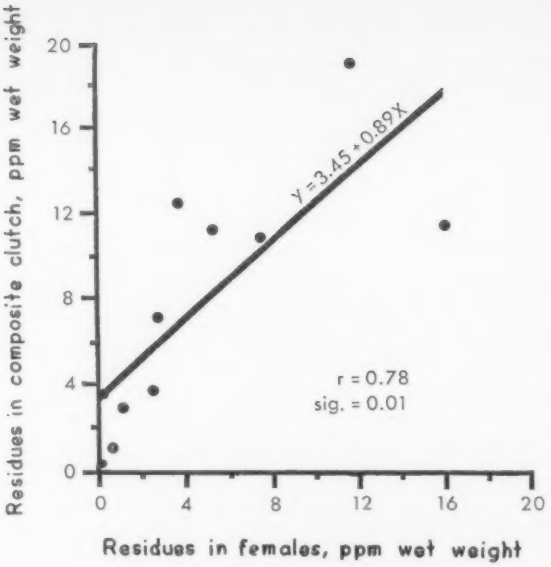

FIGURE 4. *DDE residues in female oldsquaws and their clutches collected at Rankin Inlet and Eskimo Point, Northwest Territory, June-July 1971*

13.7 ppm in a juvenile male, and the lowest in an adult was 16.4 ppm. Of the 20 birds collected as breeding pairs, only 4 (2 pairs) had DDE residues higher than 14 ppm, and 11 of the 20 paired oldsquaws had less than 5 ppm DDE. In addition, the breeding pair data exhibit a much wider spread between the highest and lowest DDE residues recorded (37×) than between mature oldsquaws collected on Lake Michigan during May 1971 (4×) or for the entire period from December to May (11×). This suggests that the breeding pair population collected at Rankin Inlet may have wintered in more than one area and these subpopulations were exposed to differing levels of DDE contamination.

Females with Clutches—Female oldsquaws with their clutches were collected approximately 1 month after the breeding pairs had been obtained. Averages for DDE and endrin in the females with clutches were lower than for the paired females but these differences were not significant (Table 7; $P>0.05$). Residues in composite clutches averaged higher than in the females that laid the clutches ($P<0.05$); laying or incubating females averaged 4.7 ppm DDE, and their clutches contained an average of 7.6 ppm DDE. PCB residues averaged 24 ppm in the females and twice that amount in the eggs. For all three compounds, average residues in the clutches were higher than in the laying or incubating hens, paired hens, or paired males.

Figure 4 indicates a strong relationship ($r = 0.78$; $P<0.01$) between DDE present in the clutch and the female collected with that clutch. Females with high levels of DDE tended to lay clutches with high DDE content, whereas hens with low DDE residues tended to transfer little of that compound to the egg.

The eggshell thickness index discussed by Ratcliffe *(26)* was used to measure differences between eggs reported here and those eggs collected before the heavy use of DDT. The mean eggshell thickness index for 106 oldsquaw eggs collected before 1947 was 1.55 ± 0.01; the average eggshell index for 75 eggs collected in 1971 was 1.48 ± 0.02. This was a decline of 4.5% ($P<0.005$).

Females with Broods—Three broods with accompanying females were collected approximately 1 month after incubating and laying females had been collected. Average residue content in females with broods was lower than in females collected previously on nests (Table 7), but again the differences were not significant. The limited brood sample did not show a positive correlation between duckling residues and the female as did the clutch and female data, but average residues were lower in broods than in clutches.

Other Collections—On August 8, 1971, authors collected five adult females molting on the sea near the mouth of the Diana River, NWT. These females were part of a group of approximately 200 other oldsquaws, all adult females. Average organochlorine pesticide and PCB residues in this sample fell between those in females with broods collected 1-2 weeks earlier and those in females with clutches collected 1 month earlier (Table 7). Since no broods were observed in the flock, either these females had not bred that year, or they had lost their clutches to predation. By this time, the ovaries of the molting females had regressed to normal winter sizes recorded on Lake Michigan.

A few yearling males were observed throughout the summer, and they were collected when the opportunity arose (Table 7). Average residues in these males were similar to those of adults collected in the Arctic but lower than residues in juvenile males collected on Lake Michigan in May 1971.

Little food was found in the gullets of recently arrived breeding pairs and females with clutches. The liver and entire gastrointestinal tract were quite small, indicating reduced use. Esophageal contents from birds collected in the late summer provided material for comparing organochlorine residues in the Arctic summer foods of oldsquaws with those found in the primary food of oldsquaws wintering on Lake Michigan. PCB residues were the highest (Table 8), but only one-tenth as high as the average residues found in Lake Michigan food samples. No endrin was detected. These data indicate that there is some contamination of the oldsquaw food base in the Arctic habitat sampled, but the level is much lower than the exposure level on Lake Michigan during the winter.

OTHER WINTERING AREAS

Oldsquaws were obtained from five wintering areas other than Lake Michigan. These collections were made in different years or at different times in the same year, depending on when cooperators could obtain specimens. Not all age/sex classes analyzed on Lake Michigan are represented in these samples. Consequently, only broad generalizations have been made from the data presented in Table 9.

Specimens collected on Lake Huron and Lake Ontario exhibit pesticide profiles similar to those recorded for oldsquaws from Lake Michigan; the orders of magnitude are also similar. Oldsquaws from Maine and Newfoundland averaged lower levels of each pesticide than did similar samples from the Great Lakes during the same time period, and those birds secured from Alaska in February contained the lowest levels of any given pesticide for all areas sampled. Endrin was not found in the eastern collections and occurred as a trace in one of the 10 specimens analyzed from Alaska.

In general, these data suggest a continuum in contamination of wintering oldsquaw populations. Those populations wintering in heavily polluted waters such as the Great Lakes contain high residues of PCBs and DDE. Populations that winter in coastal areas near highly developed urban and industrial centers contained lower residues. Oldsquaws wintering in an essentially pelagic environment far from major sources of pollution had low or undetected residue levels.

TABLE 8. *Mean and range of organochlorine pesticide and PCB residues in gullet samples of oldsquaws collected at Eskimo Point and Rankin Inlet, Northwest Territory, June-July 1971*

Compound	Residues, ppm Wet Weight	
	Mean (n=5)	Range
p,p'-DDE	0.02	0.006-0.060
PCBs	0.28	0.160-0.580
Endrin	0.00	

Discussion

Few studies have reported pesticides in oldsquaws or other ducks from Arctic areas. Five oldsquaws collected on February 24, 1964 from Lake Michigan had pesticide residues similar to those described here *(16)*. Low levels of DDE and PCBs were recorded in the neck fat of ducks off Greenland: oldsquaws contained 0.8-1.3 ppm, eiders *(Somateria spectabilis* and *S. mollissima)* contained 0.8-2.8 ppm; and harlequin ducks *(H. histrionicus)* contained 0.7-1.9 ppm DDE and PCBs *(3)*.

Various investigations present conflicting evidence as to whether DDE reaches an asymptote or continues to increase with continuing exposure. Dindal *(6)* found that DDE leveled off in mallard *(Anas platyrhynchos)* and lesser scaup *(Aythya affinia)*. Ludke *(23)* found that DDE peaked in *C. coturnix* if this was the only compound present, but if fed in conjuction with dieldrin, neither compound reached equilibrium by the end of the 56-day experiment. Data collected here indicate that DDE continued to accumulate in oldsquaws as long as they remained on Lake Michigan.

Studies on mallards and black ducks have shown that a diet containing 10 or more ppm DDE (dry weight) increased the incidence of cracked eggs, thin shells, and embryo and duckling mortality. In mallards, the ingestion of 40 ppm DDE caused the same phenomena, and they occurred earlier than in the ducks that had ingested lower levels of DDE *(13)*. Black duck females dosed with 10 ppm and 30 ppm DDE (dry weight) produced similarly detrimental effects, and the eggs averaged 46.3 ppm and 144.1 ppm DDE, respectively *(22)*. These data are similar to those reported for the brown pelicans *(Pelecanus occidentalis)* on Anacapa Island, California, when this species was in serious reproductive trouble *(21)*.

Data concerning detrimental reproductive effects in the oldsquaws can be compared with the above studies on mallards and black ducks only indirectly because residues in females that produced the observed levels in eggs were not determined and since dosages less than 10 ppm DDE were not used, the point at which statistically measurable detrimental effects appear in waterfowl is unknown. The eggs collected in the Arctic in this study had relatively thick shells; the thickness index averaged only 4.5% less than in eggs collected before the heavy use of DDT. Eggs collected in 1971 averaged only 7.6 ppm DDE. These data suggest that the species is not in any serious reproductive

TABLE 9. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected from wintering ground in the United States and Canada, other than Lake Michigan, 1971-73*

Date	Age/Sex Class	*n*	Residues, ppm Wet Weight		
			p,p'-DDE	PCBs	Endrin
LAKE HURON (SAGINAW BAY)					
11 May 1971	Mature male	2	19.4 7.4-31.3	62 47-77	0.3 0.2-0.4
	Mature female	1	21.9	50	0.4
	Juvenile male	2	8.2 5.7-10.7	21 20-22	0.2 0.2-0.3
	Juvenile female	0	-	-	-
LAKE ONTARIO (PRESQU'ILE POINT)					
23 Nov.-13 Dec. 1972	Mature male	1	5.7	32	<0.1
	Mature female	4	1.9 0.6-2.2	21 3-35	<0.1 <0.1-<0.1
	Juvenile male	4	1.2 0.9-2.2	9 3-20	0.1 <0.1-0.2
	Juvenile female	4	1.6 1.0-2.5	9 5-15	0.1 <0.1-0.3
MAINE (PENOBSCOT BAY)					
11 Mar. 1972	Mature male	6	1.7 0.4-5.5	9 2-25	ND -
	Mature female	5	0.6 0.3-0.8	4 2-5	ND -
	Juvenile male	0	-	-	-
	Juvenile female	0	-	-	-
NEWFOUNDLAND (ST. JOHN'S)					
1-5 May 1971	Mature male	2	0.3 0.2-0.4	1 1-1	ND -
	Mature female	2	0.2 0.2-0.3	1 1-1	ND -
	Juvenile male	2	0.2 0.2-0.2	1 1-1	ND -
	Juvenile female	2	0.1 0.1-0.1	1 1-1	ND -
ALASKA (POINT BAKER)					
18-23 Feb. 1973	Mature male	4	0.1 0.1-0.2	1 <1-1	Tr ND-Tr
	Mature female	4	0.1 0.1-0.2	<1 <1-<1	ND -
	Juvenile male	1	<0.1	<1	ND
	Juvenile female	1	0.1	<1	ND

NOTE: ND = not detected = no peak observed; Tr = trace = <0.005 ppm;— = no specimens collected. All values were used in computing means; trace values were used as one-half the stated "less than" value and ND was used as zero.

trouble. However, few females collected in the Arctic had DDE levels similar to those in ducks collected on Lake Michigan in May. If it is assumed that detrimental reproductive effects are similar in black ducks and oldsquaws with similar body burdens, and that the data in Figure 4 correlating DDE in the female and clutch are similarly proportional for hens with higher concentrations of DDE, then the equation of the line predicts that it would take a female oldsquaw with approximately 48 ppm DDE in her body to produce a clutch averaging 44 ppm DDE. Adverse

effects on black duck reproduction have been recorded at this level *(22)*. Since one of the four adult female oldsquaws collected on Lake Michigan in May contained 58.3 ppm DDE, these data suggest that some of the oldsquaws wintering on Lake Michigan may have reproductive problems.

PCBs were relatively high in Lake Michigan oldsquaws. Several studies have shown seriously adverse effects of PCBs on reproduction in chickens, but up to 50 ppm PCBs have not caused any measurable reproductive effects or eggshell thinning in mallards *(14, 28)*.

Body fat stores various organochlorine compounds and reduces the possibility of immediate toxic effects *(32)*. If lipid stores remain high, these chemicals can be eliminated by gradual mobilization and excretion. Studies on changes in oldsquaw body weights indicate that body lipids in Lake Michigan oldsquaws are highest (19-24 percent of carcass weight) in December-January when the pesticide body burden is relatively low, and also in May (23-25 percent of carcass weight) just before migration when pesticide body burdens are relatively high *(7, 24)*. There are two periods when oldsquaws have relatively little fat. During April, oldsquaws on Lake Michigan average only 7-9 percent lipids, and in early August, adult females with newly hatched broods averaged less than 2 percent fat. Residues in brain tissue were not measured, but if high concentrations of pesticide are present and mobilized when lipids are reduced, then April and August could be critical months. Massive mobilization of lipids and the stored pesticides can be similar to the experimental situation in which a high acute oral dose of DDT induced 25 percent thinning in mallard eggshells *(30)*. If, for lack of food or other physiological reasons, oldsquaws do not accumulate high lipid levels before migration from Lake Michigan, stored residues could be lethal to the breeding female as well as to the developing embryo.

Studies on food habits of the oldsquaw indicate that the diet of this species is 99 percent animal matter and that this is primarily an amphipod, *Pontoporeia affinis (7,25)*. Studies relating pesticide concentration to diet have generally shown that animal feeders and those species high on the food chain exhibit high concentrations of organochlorine compounds *(6, 8)*. Oldsquaws wintering on Lake Michigan between 1969 and 1972 were on a continuous diet of amphipods containing an average of 1.27 ppm DDE, and adult female oldsquaws averaged 32.1 ppm DDE just before spring migration in 1971. Hickey et al. *(16)* found that DDE residues in *P. affinis* varied between 0.19 ppm and 0.24 ppm in July 1964. This is considerably lower than the estimates of average levels in amphipods reported here, but their calculated concentration factor of 15× in February between *P. affinis* and oldsquaws is within the range of 4× (December) to 21× (May) reported in this paper.

Even though oldsquaws and whitefish in Lake Michigan eat the same organisms and are, therefore, on the same trophic level, residues reach much higher levels in the birds than in the fish. DDE residues in whitefish reported by Hickey et al. *(16)*, Reinert (DDE-TDE-DDT) *(27)*, and the Wisconsin Department of Natural Resources (WDNR; unpublished data) were 3.00, 0.78, and 0.64 ppm, respectively, whereas DDE averaged 6.1 ppm for all oldsquaws collected in December 1970, and 25.6 ppm in May 1971. PCBs averaged 4.3 ppm in the Wisconsin whitefish, and December and May averages for all oldsquaws were 12.5 ppm and 67.2 ppm, respectively. It seems reasonable to expect higher levels in birds than in fish that eat the same food, because birds are warm-blooded and would require more energy than would fish.

The relationship of age and sex with the accumulation of pesticides in avian species has been little studied. Young nonbreeding birds show only minimal sex-dependent differences in susceptibility, but this might not hold for mature birds during the breeding season *(31)*. Experimental LD_{50} values for DDT have been shown to vary with age in mallards *(9)*, and DDE has appeared to be higher in male than in female mallard wings although the differences have not been significant *(15)*. It has been hypothesized that the greater resistance of adult females versus adult males to pesticide toxicity was partly a result of the females' ability to excrete pesticides through their eggs *(9, 17)*. Early winter differences in DDE residues among different age and sex classes of oldsquaws, as well as the positive relationship between DDE in the female and clutch, suggest that breeding female oldsquaws can eliminate pesticides through the egg and return to wintering areas with a lower body burden than can nonbreeding females or males that presumably have bred.

A number of studies have pointed out regional differences in the pesticide burdens of various avian species and populations. Because of differences in age, dietary habits, and possibly winter migration to more heavily polluted areas, the amount of polychlorinated hydrocarbons may vary from one species to another *(3)*. Keith *(19)* summarized regional differences in pesticides when he stated: "Early results show truly astonishing regional within-species variations in residue loads, and these require explanation presumably in terms of local pesticide use patterns and local food preference." The nationwide pesticide monitoring program conducted on mallard and black duck wings also showed State and regional differences in DDE and PCB levels *(12, 15)*. Data from the present study concerning the Great Lakes as well as Atlantic and Pacific coastal areas support these conclusions concerning regional differences. The relatively low levels in oldsquaws wintering in estuaries or pelagic areas confirm an early prediction by Butler and Springer *(4)* that ". . . because of rapid dilution rates, high pesticidal concentrations are not to be anticipated in estuaries."

Only about 200 oldsquaws have been banded in North

America outside Alaska (unpublished data, Migratory Bird Population Station, Laurel, Maryland, 2). Consequently, little is known of the migratory movements of this species. Oldsquaws primarily winter on the Great Lakes and in coastal areas of the northern United States and southern Canada *(18)*. Since marked differences in pesticide residues appear in oldsquaws collected from these areas, it may be possible to use characteristic pesticide profiles of these compounds in place of leg bands to trace movements of the birds. Thus the wintering grounds of different breeding birds might be determined, at least approximately, by comparing the amounts, kinds, and ratios of pesticides found in various populations.

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Mercury, Cadmium, Lead, and Arsenic in Sediments, Plankton, and Clams from Lake Washington and Sardis Reservoir, Mississippi, October 1975-May 1976 [1]

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ABSTRACT

Mercury, cadmium, lead, and arsenic concentrations were measured in clams, plankton, and sediments from Lake Washington, Washington County, an oxbow lake off the Mississippi River, and Sardis Reservoir, Lafayette and Panola Counties, a flood control reservoir on the Little Tallahatchie River. Mercury, cadmium, and lead were measured by atomic absorption spectrophotometric methods; arsenic was measured by the silver diethyldithiocarbamate method. Correlation coefficients for concentrations of metals in the sediments, plankton, and clams were derived. Mean concentrations for all metals were higher in plankton and clams than in sediment, although levels were only slightly higher in clams than in sediments. Plankton contained the highest levels of all four metals. There were no significant correlations between the metals for sediments or clams. A t-test used to compare concentrations of the four metals in the sediments of the two bodies of water showed a significant difference only for arsenic concentration. A high correlation of mercury between plankton and clams indicates the food web relationships. There was also a positive correlation of cadmium between sediments and clams, and a correlation of arsenic between sediments and plankton. Comparisons between clam size and trace metal concentrations showed higher concentrations of mercury in the smaller clams.

Introduction

During the past 23 years, trace metal pollution in aquatic systems has been researched extensively. This was brought about by the tragedy in Minamata City, Japan in 1953 in which 41 persons died of what was subsequently shown to be mercury poisoning (*19,15,16*). Effluent from a factory where $HgCl_2$ was used as a catalyst in the production of vinyl chloride was discharged into Minamata Bay; as a result, fish and shellfish from the bay, which were a major source of food for the villagers, accumulated high levels of mercury. Since the Minamata City tragedy and the realization of the toxicity of low concentrations of heavy metals, data concerning mercury pollution have increased, and other elements such as lead, cadmium, and arsenic are being investigated.

The occurrence of trace elements in the biosphere at higher levels than in the hydrosphere has been recognized for some time (*23, 25, 6*), but the order of magnitude among organisms varies considerably. The mechanisms by which trace metals are concentrated by organisms are still not fully understood, but there is evidence that accumulation occurs through the food chain (*23, 12*).

Because of their potential toxicity to humans and their persistence in the environment, certain heavy metals which accumulate through the food chain are of particular interest. The magnifying effects at the higher trophic levels make higher organisms particularly vulnerable to these elements. Radionuclides, chlorinated hydrocarbons, and trace metals pose a serious problem in the aquatic environment because they are concentrated from the water into sediments and the food web (*18*).

It has been shown that Pelecypoda feed on plankton (*1, 20*) and can selectively concentrate pollutants against a gradient (*23*). Although certain materials are sorted before entering the stomach, all particles, including great quantities of sediment, are retained by the gills (*8*); stomach contents also contain detectable quantities of sediments (*13*).

In this study, mercury, cadmium, lead, and arsenic were measured and compared in sediments, clams, and plankton from Lake Washington and Sardis Reservoir, Mississippi, to study food web relationships.

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Description of Sardis Reservoir and Lake Washington

Sardis Reservoir is part of a long-range plan for flood control of the Yazoo River Basin. The dam and appurtenances were developed by the U.S. Army Corps of Engineers in 1940. At full capacity, the reservoir has a surface area of 237 km^2 with an average depth of 5.5 m. Water level fluctuates considerably as water is periodically drained; at the lowest water level, the conservation pool is about 43 km^2.

Lake Washington is an oxbow off the Mississippi River, located 32 km south of Greenville, Mississippi. It is a large shallow lake occupying 20 km^2 in Washington County. One-fourth of the lake contains cypress trees in about 1 m of water. Water level fluctuates no more than 2 m; the high level occurs in the spring and the low level occurs in the fall.

Fish from Lake Washington contained pesticide residues at concentrations above the action level of the Food and Drug Administration, U.S. Department of Health, Education and Welfare (9), thus the lake is presently closed to commercial fishing and the public has been warned against consuming fish from the lake.

Materials and Methods

Samples were collected monthly from October 1975 through May 1976. Three stations on each body of water were sampled each month; a fourth station was sampled intermittently (Fig. 1, 2). Samples of the upper 6 cm of sediment were collected as near as possible to the sites of the plankton samples.

Clam, sediment, and plankton samples were placed in appropriate containers and frozen on the day of collection to ensure that no trace metals would be lost to volatilization.

Plankton samples were collected by using a pump and hose apparatus and were concentrated by using a No. 25 plankton net. The pump consisted of five individual marine and camping 12-volt dc pumps arranged radially symmetrical on a 3.1-mm-thick Plexiglas disk 40 cm in diameter.

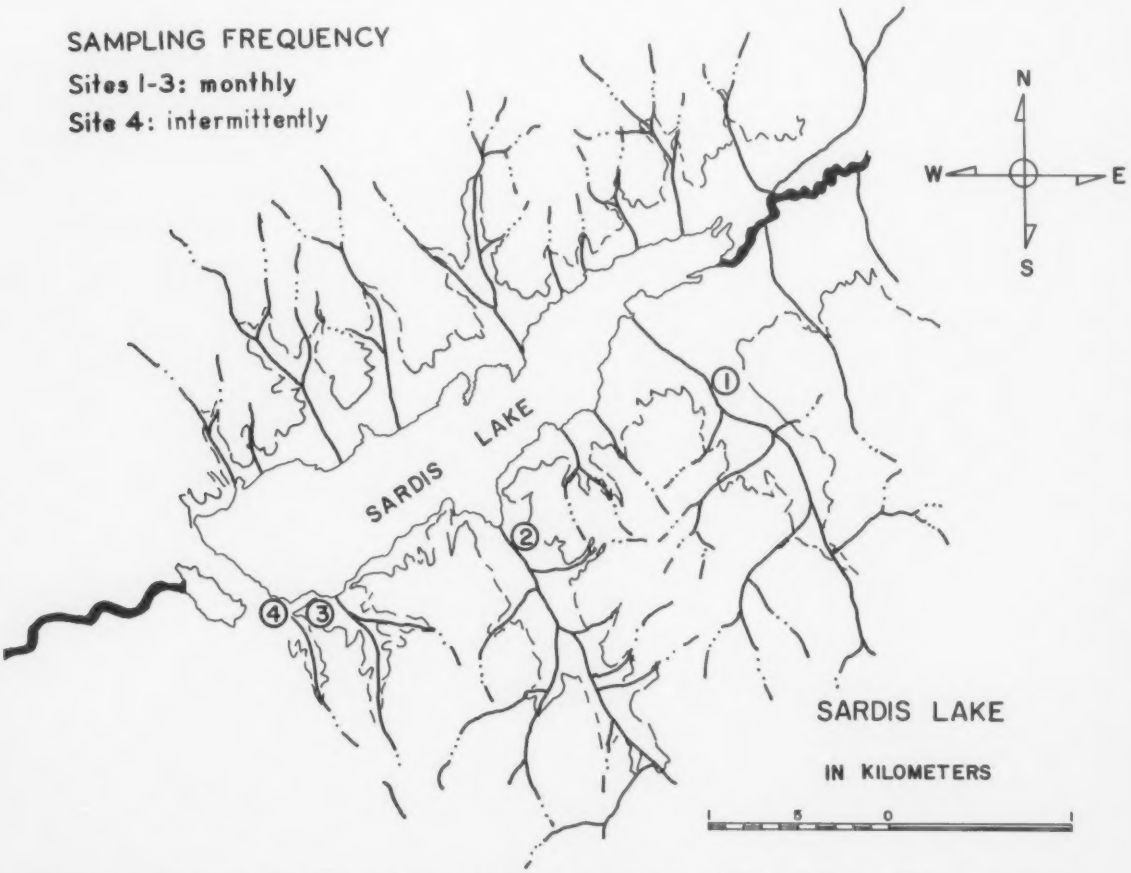

FIGURE 1. *Sardis Reservoir (34° 30' north latitude, 89° 40' west longitude), showing selected sites for collecting sediments, plankton, and clams for heavy metal determinations*

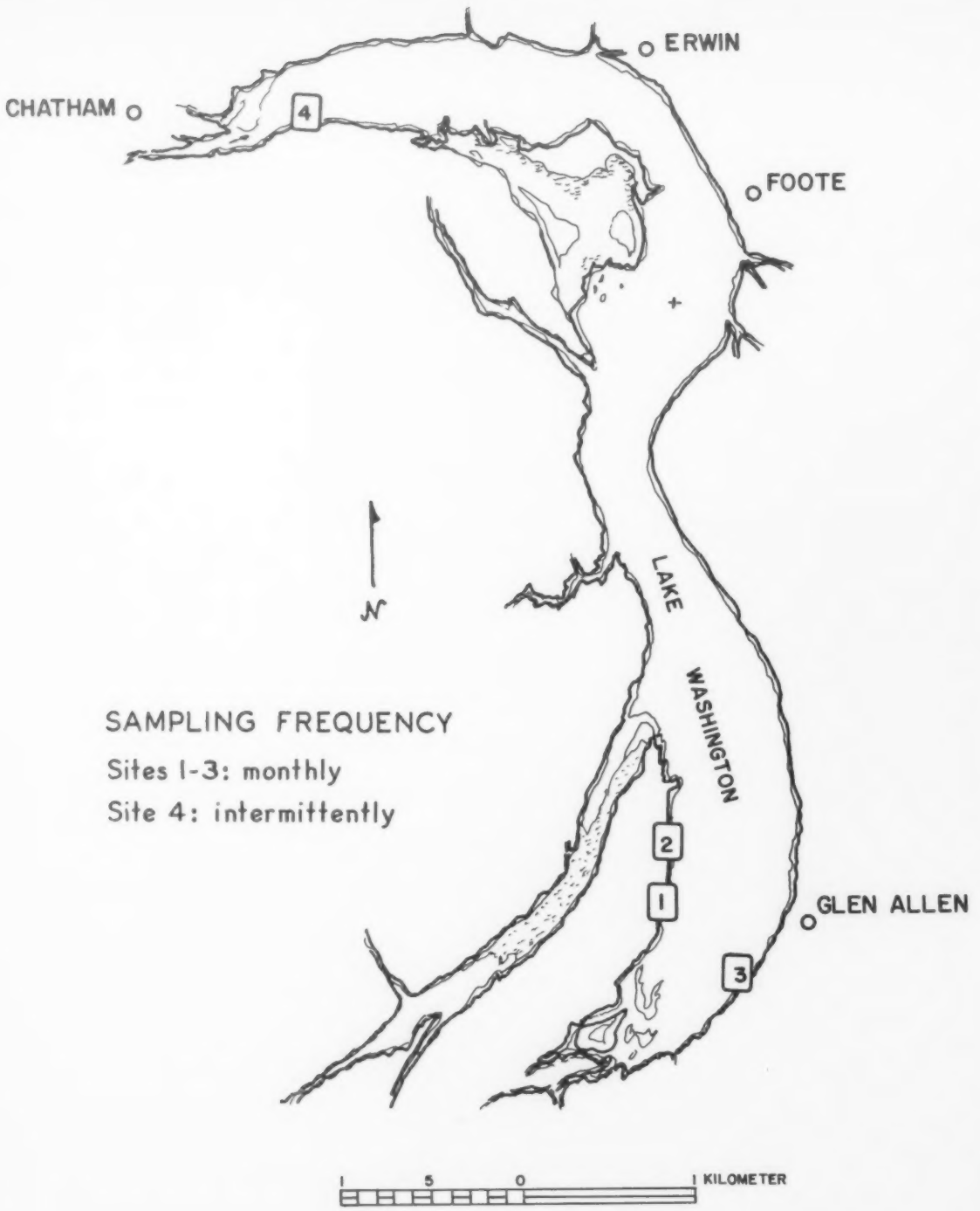

FIGURE 2. *Lake Washington (33° 05' north latitude, 91° 05'* west longitude), showing sites selected for collecting sediments, *plankton, and clams for heavy metal determinations*

Incurrent holes of the pumps were located 11.7 cm from the periphery of the disk. Another disk of the same size was placed 1.9 cm below the first disk and mounted on feet which were 9.5 cm long. Thus plankton samples were obtained 10.0 cm above the bottom. Because the disk was parallel to the bottom during operation, the water was pulled in horizontally. All pumps were joined in the center to a 3.8-cm hose which led to the surface.

The plankton pump was calibrated before and after each run in the time required to fill a container with 20 liters of water; then the amount of water filtered by the net was calculated.

A compound microscope equipped with a draw tube, 10×-ocular, and 16-mm objective was used in combination with a Whipple ocular micrometer for plankton enumeration. An aliquot was placed in a Sedgwick-Raftar counting cell, and field counts were made. For filamentous forms, a length of 300 μ was considered a unit. All other forms, including individual cells and colonies, were counted as single units. Plankton counts are summarized in Table 1.

Clams were collected by hand with a garden rake or by a dredge constructed after a model from Burch (*7*).

Samples were analyzed for mercury by the flameless atomic absorption technique developed by Hatch and Ott (*14*). Samples were digested with 5 ml of concentrated nitric acid and 5 ml of concentrated sulfuric acid for 12 hours, and then reduced with 5 ml of potassium permanganate and 5 ml of potassium persulfate for 12 hours. The reaction flask was placed in the closed system, and stannous sulfate in a sodium chloride-hydroxylamine medium was added to reduce the mercury to its elemental state. Once maximum absorbance was obtained, the mercury vapor was vented to the hood. Standards were analyzed by the same procedure. The standard values were statistically corrected by the method of least squares (*2*) and were used to obtain a calibration curve. A Beckman Model 1301 atomic absorption spectrophotometer equipped with a 10-inch potentiometric recorder was used for the analysis.

Sediment samples for lead, cadmium, and arsenic measurements were dried in a vacuum oven at 105°C for 24 hours. Plankton samples were dried only for arsenic determinations.

In order to use larger sample sizes of plankton, mercury, cadmium, and lead analyses were all conducted on the same sample. Samples were first analyzed for mercury, and then filtered and prepared for cadmium and lead analyses by the method of additions (*22*). Blanks were used to compensate for contamination by chemicals. Final results were obtained by the method of least squares and are reported as ppm wet weight.

Clams were also analyzed for cadmium and lead by the method of additions to eliminate matrix effects. Clam soft tissues were digested with nitric acid on a hot plate until dry. Charring of samples was minimized by adding more nitric acid. After each sample cooled, 5 ml of concentrated hydrochloric acid was added. Samples were then diluted to 25 ml with distilled water, and filtered. Five-ml aliquots were used for the method of additions.

Sediment samples for cadmium and lead analyses were digested in nitric acid to dryness on a hot plate. The residue was dissolved in concentrated hydrochloric acid, diluted to volume, and aspirated into the atomic absorption apparatus. Absorbance was compared to a standard curve obtained by the method of least squares. Results are expressed as ppm wet weight.

For arsenic analysis, various combinations of acids were tried for digestion of samples, but a 4:1:1 ratio of nitric, sulfuric, and perchloric acids (*10*) provided the best results. This mixture was added to the sample and heated until fumes of SO_3- appeared. The sample was allowed to cool and then 5 ml of concentrated hydrochloric acid was added and diluted to 35 ml with distilled water The samples were then subjected to arsine generation as described by the American Public Health Association (*2*). Standards were analyzed with each set of samples. Percent transmittances obtained from the spectrophotometer were converted to absorbance to obtain a working curve. The method of least squares was used to plot a straight line from which concentrations could be obtained. Results are expressed as ppm wet weight.

Methods, instrumentation, sensitivities, ranges, mean recoveries, and total number of analyses are presented in Table 2.

Concentration data for the four metals were arranged in three groups: one group of sediment data only (N=53, where N represents sample size), one group of sediment and plankton data (N=25), and one group of sediment, plankton, and clam data (N=19). Correlation coefficients and a *t*-test were run on each group to ascertain relationships and significant differences between the two bodies of water.

TABLE 1. *Ranges and means for net plankton counts from Sardis Reservoir and Lake Washington, Mississippi*

	Net Plankton Counts	
Statistic	Sardis Reservoir	Lake Washington
Range	493,083 - 1603	1432 -45
Mean	106,495	1308
N	21	23

TABLE 2. *Instrumental and analytical parameters for determining mercury, cadmium, lead, and arsenic in sediments, plankton, and clams*

Parameter	Metal			
	Hg	Cd	Pb	As
Method	Flameless AAS	Flame AAS	Flame AAS	Silver diethyldithiocarbamate
Instrument	Beckman Model 1301	Perkin-Elmer Model 303	Perkin-Elmer Model 303	Spectronic 20
Sensitivity, ppm	0.0005	0.20	0.40	0.5
Range, ppm	0.0005-0.020	0.05-5.0	4-40	1-10
Mean recovery (percent)[1]	97.0	99.8	101.6	76.5
Number of analyses	275	203	203	284

[1] Data are not corrected for percent recoveries.

TABLE 3. *Mean concentration of heavy metals in sediment, plankton, and clams from Lake Washington and Sardis Reservoir, Mississippi*

Sample	Metal			
	Hg	Cd	Pb	As
Sediment	0.0481	0.3614	8.4847	2.99
Plankton	0.4551	14.4587	501.5964	21.74
Clams	0.1073	0.2123	11.5685	0.41

NOTE: Concentrations in ppm wet weight. N=19.

Results and Discussion

A *t*-test on the group containing 19 cases with complete samples, i.e., sediment, plankton, and clam data, showed no significant difference between the two bodies of water. These results were pooled to obtain one value to show any food web relationships (Table 3). In all cases, mean concentrations were higher in the plankton and clams than in the sediment.

Mercury may have been biomagnified, with increased concentrations from sediments to plankton and clams. The higher levels in plankton compared to clams may be due to the trophic conditions of the water (*17*). According to Smith and Green (*25*), water must be severely polluted before mercury levels increase in clams. The habitats in the present study are relatively uncontaminated; this could explain why the clams contained only slightly higher levels of mercury than did the sediments.

Frazier stated that cadmium uptake by oysters is direct and independent of the food web and that clams also have been used as indicators of cadmium levels in their environment (*11*). This suggestion of bioconcentration is illustrated by the similar concentrations of cadmium in sediments and clams. Plankton are known to accumulate cadmium; this is supported by the results of the present study.

Background levels of lead have been widely reported, but no single value is universally accepted. Uptake of lead by plankton through surface adsorption has been demonstrated (*21*), and this is shown in the present study. Remarkably uniform concentrations of lead in mussels have been reported (*4*) and are representative of the particular environment in which they are found (*23, 5, 24*). Clams in Sardis Reservoir and Lake Washington appear to concentrate lead only to levels barely exceeding those in sediment.

Plankton are known to accumulate and magnify arsenic to concentrations above that found in the water (*36*). The large amounts of arsenic in plankton in the present study illustrate this point. Arsenic data for Mollusca are scarce, and higher organisms are not known to accumulate arsenic (*26*). The low value in this study indicates little concentration of arsenic by Mollusca.

A *t*-test with only sediment values for the two bodies of water showed a significant difference between arsenic in the sediments: 3.6 ppm in Sardis Reservoir and only 2.4 ppm in Lake Washington.

When arsenic enters the aquatic environment, it settles and accumulates in the sediments (*26*); levels in sediments have been used for water appraisal (*3*). The difference between the two lakes may be explained by the large amount of runoff which Sardis Reservoir receives.

With sediment data (Table 4), a correlation matrix used to ascertain relationships among the four metals showed one significant value ($P > 0.50$): mercury with date of sample. This may indicate some seasonal cycle. Nonsignificant values for the rest of the matrix indicate no correlations between the metals in the sediments.

A *t*-test for heavy metals in clams from the two lakes revealed that there were no significant differences between the two groups. The absence of significant differences suggests that the mode of uptake of the metals examined for the clams from the two bodies of water must be similar.

TABLE 4. *Correlation matrix for heavy metals in sediment*

Metal	Hg	Cd	Pb	As
Hg	1.00			
Cd	-0.16	1.00		
Pb	0.15	0.23	1.00	
As	0.32	0.06	0.34	1.00
Date	0.61	-0.30	0.06	0.29

NOTE: N=53.

TABLE 5. *Correlation matrix for heavy metals in clams*

Metal	Hg	Cd	Pb	As
Hg	1.00			
Cd	-0.09	1.00		
Pb	-0.22	-0.09	1.00	
As	-0.10	-0.00	0.02	1.00
Date	0.33	-0.19	0.02	-0.24

NOTE: N=19.

TABLE 6. *Correlation matrix for heavy metals in plankton*

Metal	Hg	Cd	Pb	As
Hg	1.00			
Cd	0.98	1.00		
Pb	0.98	0.96	1.00	
As	-0.12	-0.12	-0.06	1.00
Date	-0.28	-0.26	-0.24	0.07

NOTE: N=25.

TABLE 7. *Correlation coefficients for heavy metals in sediments, plankton, and clams*

Material	Hg	Cd	Pb	As
Sediment/plankton	-0.08	-0.17	-0.06	0.45
Plankton/clams	0.45	-0.20	-0.12	0.31
Sediment/clams	-0.39	0.16	-0.07	0.13

NOTE: N=19.

Concentrations of heavy metals in clams from the two bodies of water were then pooled for correlation analysis of the heavy metals. The correlation matrix (Table 5) for the clams showed no significant values, indicating that there were no reciprocal relations between the metals in the clams.

The correlation matrix for heavy metals in the plankton (Table 6) shows some high correlations between cadmium and mercury, cadmium and lead, and mercury and lead. These unexpected high values are due to experimental design: all three analyses were conducted on the same sample. A *t*-test on this group of data revealed no significant difference in metal content in the plankton from either body of water.

The correlation coefficients for heavy metals in sediments, plankton, and clams (Table 7) further illustrate some trends. A high correlation of mercury between plankton and clams indicates the food web relationships of the two. The positive correlation of cadmium between the sediments and clams indicates their relationship as mentioned earlier. Lead shows no particular trends with this matrix. Arsenic displays the sediment/plankton relationship as discussed earlier.

Data were regrouped to test for possible relationships of metal concentration with the various species of clams (Table 8). Eight species were analyzed: *Corbicula manilensis* (Philippi), *Proptera purpurata* (Lamarck), *Anodonta grandis corpulenta* (Say), *Anodonta suborbiculata* (Say), *Anodonta grandis* (Say), *Carunclina parva* (Barnes), *Quadrula pustulosa* (Lea), and *Lampsilis anodontoides* (Lea). Of these, *A. grandis*, *Q. pustulosa*, and *L. anodontoides* came from Lake Washington. *A. g. corpulenta*, *C. manilensis*, and *P. purpurata* came from Sardis Reservoir. *A. suborbiculata* and *C. parva* were obtained from both lakes. This provided an even distribution of species comparison.

Smaller clams, i.e., *C. parva*, *Q. pustulosa*, *C. manilensis*, and some *L. anodontoides*, contained higher levels of mercury than did the larger clams. Since smaller organisms have higher surface-to-volume ratios, they would be expected to have greater uptake.

Cadmium appeared to be concentrated in approximately the same amounts regardless of species.

Clams from Lake Washington contained higher levels of lead than did *A. g. corpulenta*, *C. manilensis*, or *P. purpurata* from Sardis Reservoir. These higher levels may be due to the richer trophic condition and the age of Lake Washington.

Arsenic levels were consistently low in all species, reinforcing the theory of little or no arsenic accumulation by Mollusca.

Summary

A significant correlation of mercury in the sediments with date of samples indicates some seasonal trends.

The *t*-tests revealed no significant differences between Lake Washington, an oxbow lake, and Sardis Reservoir, a flood control reservoir, with respect to the metals in the

TABLE 8. *Ranges and means of heavy metals in clams from Lake Washington and Sardis Reservoir, Mississippi*

Species of clam	Residues, ppm Wet Weight			
	Hg	Cd	Pb	As
Proptera purpurata				
Range	0.000-0.034	0.000-0.800	0.000-6.910	0.00-0.72
Mean	0.017	0.311	3.396	0.19
Carunculina parva				
Range	0.000-0.463	0.000-1.170	0.000-89.262	0.00-1.03
Mean	0.043	0.288	9.433	0.22
Anodonta suborbiculata				
Range	0.000-0.012	0.031-0.380	0.000-1.023	0.00-1.93
Mean	0.001	0.175	0.544	0.30
Anodonta grandis corpulenta				
Range	0.000-0.011	0.000-0.521	0.000-1.167	0.00-0.36
Mean	0.005	0.120	0.330	0.11
Quadrula pustulosa				
Range	0.000-0.432	0.000-0.243	0.000-31.857	0.00-1.39
Mean	0.087	0.086	8.777	0.36
Lampsilis anodontoides				
Range	0.000-0.149	0.000-0.212	0.585-6.360	0.00-0.54
Mean	0.058	0.160	3.313	0.06
Anodonta grandis				
Range	0.000-0.166	0.000-0.547	0.000-33.847	0.00-0.32
Mean	0.008	0.230	5.082	0.10
Corbicula manilensis				
Range	0.000-0.218	0.000-0.235	0.000-1.429	0.30-0.33
Mean	0.063	0.151	0.614	0.31

biosphere, although there was a significant difference in arsenic content of the sediments.

Small clams contained higher levels of mercury than did large ones.

Clams from Lake Washington contained higher levels of lead than did Sardis clams.

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RESIDUES IN WATER

Residues of Polychlorinated Biphenyls, DDT, and DDT Metabolites in Pennsylvania Streams, Community Water Supplies, and Reservoirs, 1974–76 [1]

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ABSTRACT

Pennsylvania streams, community water supplies, and open water reservoirs were analyzed for PCB mixtures, DDT, and DDT metabolites. Streams were sampled in 1974 and again in 1975. Only 4 of 19 stream locations were contaminated. Maximum concentrations in community supplies of Aroclor 1242 (2 locations), Aroclor 1254 (3 locations), and ΣDDT (2 locations) were 350, 260, and 620 ng/kg, respectively. Of the 110 community water supplies sampled in 1975 and 1976, only 7 contained residues. Maximum concentrations in community supplies of Aroclor 1242 (4 locations) and ΣDDT (2 locations) were 460 ng/kg and 75 ng/kg, respectively. The seventh contained 0.7 ng/kg of dieldrin. None of the three open water reservoirs contained detectable residues of the compounds of interest.

Essentially no correlation was found between PCB and DDT analogs in streams and those in fish from streams which had been sampled at similar locations in a related study in 1976.

Introduction

Polychlorinated biphenyls (PCBs) and residual DDT and DDT metabolites (ΣDDT) are ubiquitous environmental contaminants not only in the United States but in other parts of the world (*8*). Contamination has spread from localized points to broad areas by many modes of transport, one of which is water.

The history of pesticide analysis in water can be traced to the 1950s in publications of the Public Health Service, U.S. Department of Health, Education, and Welfare, and the Geological Survey, U.S. Department of Interior (*7*). The Geological Survey began monitoring under the National Water Quality Network in 1965 and focused on the analysis of western streams (*21*).

Only within the past few years has there been a directive to study the nationwide contamination of water systems by PCBs and DDT. Federal drinking water limits for chlorinated hydrocarbons were first released in 1973 (*17*). In 1975, the U.S. Environmental Protection Agency (EPA) published national interim primary drinking water standards pursuant to requirements set by the Safe Drinking Water Act (Pub. L. 93–523) (*23*). Requirements of this act included the study of PCB contamination for the first time. In 1977, EPA set toxic effluent standards for DDT analogs (*24*) and for PCBs (*25*).

A summary of the residue studies conducted nationwide from January 1971 through June 1972 has been published (*8*). At that time, PCBs were found in the water of 12 States. Eight States were sampled moderately (25–50 samples) and three States were sampled heavily (greater than 160 samples), but Pennsylvania was sampled only twice.

Recent individual studies relating to environmental contamination of these long-lasting compounds have been reported. The occurrence of chlorinated hydrocarbon insecticides in southern Florida in 1968–72 was determined (*15*). Seasonal variations were found in residues of chlorinated hydrocarbons in the water of the Utah Lake drainage system (*4*). DDT analogs and PCBs were found in all stream tributaries to San Francisco Bay (*14*). Selected western streams frequently contained DDT analogs in 1967–71 but PCBs were found only twice in 1969–71 (*20*). Organochlorine pesticides were found in the Arkansas River in the vicinity of Tulsa, Oklahoma, in 1970–72, although no DDT analogs were found (*16*). Insecticide residues were studied (*12*) in the Tuttle Creek Reservoir ecosystem, Kansas, in 1970–71. Highly sensitive analyses

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were made for chlorinated hydrocarbon residues in the lower Mississippi River (*6*). Most samples of drinking water studied on the Island of Oahu, Hawaii, in early 1971 contained *p,p'*-DDT (*3*). In a 1969–71 study of four small drainage basins in Pennsylvania (*22*), DDT and analogs were found in water as well as soil, streambed material, and fish. In that study, PCBs were not determined.

Ground water has also been analyzed. The movement of DDT in ground water was measured in the Ogallala Aquifer in Texas (*18, 19*). In Georgetown County, South Carolina, 27 ground water wells from the lower coastal region bordered by the Atlantic Ocean were analyzed and all contained DDT in the parts per thousand range (*1*).

These reports indicate many studies of contamination in streams but few studies of community water supplies or open water reservoirs. The present study has helped to delineate the extent of contamination in community water supplies and open water reservoirs by PCB mixtures and DDT analogs. This is the first broad study of such sources in Pennsylvania. Streams were included in the study.

Analytical Technique

FIELD PROCEDURES

Stream samples were taken at Pennsylvania Water Quality Network (WQN) Stations (*9*). They were sampled at two-to-four points along a cross section or from both sides as described in the publication. Community supplies were sampled either as finished water at the main distribution point or as source water supply points. The grab samples were collected in new bottles that had been cleaned with solvent and rinsed with sample. Caps had Teflon liners. Samples were refrigerated until tested. The samples were not preserved with formaldehyde as has been described (*2*) because some batches of formaldehyde were contaminated by Aroclor 1242.

INITIAL EXTRACTION AND CLEANUP

Chemicals and laboratory equipment have been described (*13*). Water samples were extracted by adsorption to a cellulose triacetate membráne filter (*13*). Any sediment collected was included in the analysis. Occasionally several filters were required to shorten filtration time due to clogging. PCBs and DDT analogs present in water samples collected in 1974–75 were recovered from the filters by shaking once for 30 minutes with hexane and anhydrous sodium sulfate and then twice for 10 minutes with hexane alone. The combined hexane layers were concentrated, passed through a Florisil column, and analyzed by gas-liquid chromatography (GLC).

Water samples collected in 1976 were subjected to an improved recovery procedure. The filters were precleaned with ethyl ether and rinsed with distilled water before samples were filtered. Adsorbed compounds were removed with two portions of ethyl ether. The combined ethyl ether extracts were dried, concentrated, and transferred to isooctane by successive evaporations. The samples were then passed through a Florisil column and analyzed by GLC.

Control samples of laboratory double-distilled water were analyzed at regular intervals.

ANALYSIS

GLC analyses (*13*) were performed on a 5.5-foot × ¼-inch OD glass U column packed with 1.5 percent SP-2250/1.95 percent SP-2401 on 100–120-mesh Supelcoport. The column was maintained at 215°C; the nitrogen flow was 60 ml/min.

The ^{63}Ni electron-capture detector temperature was 330°C. Electrometer sensitivity was 1.6×10^{-9} amp full scale. Peaks of interest were measured with an Infotronics Model CRS–100 area integrator.

Samples which had peaks at retention times for PCBs and/or DDT and its metabolites were subjected to silicic acid column chromatography to separate PCBs from the DDT analogs: 430-mm × 7-mm ID column containing 5 g of silicic acid (Mallinckrodt, CC-7 Special), activated for 4 hours at 190°C, was used for the separations. The packing was prewashed with toluene. PCBs were eluted with petroleum ether and the DDT analogs were eluted with toluene. Samples were rechromatographed on the above GLC column, and peak identities were confirmed on a 6-foot column of 3 percent DEGS on 80–100-mesh Chromosorb WHP.

Compounds were quantitated by comparing GLC peak areas with those obtained from standards analyzed at regular intervals. PCB mixtures were quantitated by comparing the sums of the peak areas of mixture. When two mixtures of PCB types were apparent, peaks were quantitated by using the sum of the areas of nonoverlapping peaks.

STORAGE RECOVERY

Aroclor 1254 was added to double-distilled water at two levels and stored in the refrigerator with the other samples. The storage recovery samples were analyzed after 13 months; the stream and water supply samples were analyzed after 1–9 months of storage.

Average percent recoveries of triplicate analyses were:

NG AROCLOR 1254 ADDED	PERCENT RECOVERED ($\bar{x}$)
500	97
50,000	93

RECOVERIES

PCB- and DDT-spiked distilled water samples were passed through the membrane filter and recoveries were determined after hexane extraction of the filter disks. Complete details of this procedure are described elsewhere (*13*). This

scheme was used for the analysis of samples collected in 1975. The summary of triplicate analyses at three spiking levels is:

Compound	μg Added	Percent Recovered (±SD, n=9)
Aroclor 1242	0.40, 2.0, 10	40 (±14)
Aroclor 1254	0.60, 3.0, 15	26 (±16)
p, p'-DDE	0.02, 0.1, 0.5	24 (±14)
p,p'-DDT	0.04, 0.2, 1.0	16 (±8.5)

The PCBs, DDT analogs, and others were recovered from samples collected in 1976 by ethyl ether extraction of the filter disks. Triplicate analyses at three spiking levels produced the recoveries below:

Compound	μg Added	Percent Recovered (±SD, n=9)
Aroclor 1242	0.40, 2.0, 10	98(±14)
Aroclor 1254	0.60, 3.0, 15	83 (± 2)
p,p'-DDE	0.03, 0.15, 0.75	72 (± 7)
p,p'-TDE	0.06, 0.3, 1.5	74 (± 9)
p,p'-DDT	0.06, 0.3, 1.5	76 (±15)
Mirex	0.3, 7.5	105 (±12) (n=6)

The results reported in this paper were corrected for these recoveries.

Results

Analysis of Pennsylvania water samples collected in 1974–76 showed the presence of PCBs or DDT analogs only in isolated locations (Tables 1–3). Only four stream locations of 19 Pennsylvania Water Quality Network stations sampled showed the presence of these pollutants. Of the community water supplies, only six of 110 sampled areas contained these compounds; a seventh contained dieldrin. None of the three open water reservoir samples contained any of these residues.

Stream stations were sampled twice, once in 1974 and again in 1975. Most of the stream contamination was found in Philadelphia, where the Delaware and Schuylkill Rivers contained both PCBs and DDT analogs. The Delaware River contained Aroclors 1242 and 1254, and DDE, TDE, and DDT at both sampling times. The concentrations of Aroclors in the normal flow sample taken in 1974 were 70 ng/kg (ppt) and 260 ng/kg (ppt), respectively; ΣDDT (*p,p'*-DDE + *p,p'*-TDE + *p,p'*-DDT) concentration was 620 ppt. At higher flows in fall 1975, the concentrations of Aroclors and DDT analogs were reduced, respectively, to 180 ppt and 60 ppt. At the lower end of the Schuylkill River in fall 1974, the PCB concentration was 500 ppt, but in fall 1975, no PCBs were found.

Only trace quantities of the compounds of interest were found at the other WQN stations. DDT was found at the Lehigh (Easton) station, and Aroclor 1254 was found at the Monongahela (Rankin) station upstream from Pittsburgh.

The amount of stream flow may indicate a dilution effect. The Delaware samples in Philadelphia contained fewer contaminants with increased water flow; the Lehigh samples may have been affected similarly. The dilution effect was not observed in one study (*10*), however, of a uniformly treated area where DDT residues were distributed evenly over varied flow rates.

Of the community water supplies, the 1975 Lewiston sample contained 460 ppt Aroclor 1242. Subsequent sampling in 1976 of the two runs leading to the reservoir and of one location in the distribution system did not indicate any residues. Similarly, the 1975 Lock Haven sample contained a trace of Aroclor 1242, but further sampling of this reservoir and two distribution sites in 1976 showed no residues.

The same situation occurred in two communities in southwestern Pennsylvania. The 1975 Waynesburg sample contained 75 ppt ΣDDT, yet in 1976 no DDT was found in the water supplies in four different locations of that community. Trace DDT was found in 1975 in Point Marion, but in 1976 none was found in supplies in five different locations.

Two of the community water supplies sampled in 1976 contained residues. A total of 19 communities were sampled at that time. The Masontown sample contained 20 ppt Aroclor 1242, and the Pottstown sample contained a trace of Aroclor 1242. The community water supply of Centre Township, also in that area, contained 0.7 ppt dieldrin.

Sulfur was not observed as an industrial contaminant in any of these samples.

Discussion and Conclusions

PCB AND DDT LEVELS OF CONTAMINATION

Although PCBs are currently reported to be environmentally ubiquitous (*8*), they were detected in only a few locations in Pennsylvania at any significant level: three of 19 stream locations and four of 110 community water supplies, comprising a total of 5.4 percent. The nationwide study (*8*) conducted from January 1971 through June 1972 found a similar percentage of PCB contamination: 83 of 1627, comprising 5.1 percent. In the earlier study, one of two locations sampled in Pennsylvania contained PCBs. Neither study, unfortunately, was broad enough to indicate any trend in the concentration of these pollutants.

Truhlar and Reed (*22*) studied in detail DDT and DDT analogs in four Pennsylvania streams in 1969–71. ΣDDT in samples from a forest stream (Young Womens Creek near Renovo), a farm stream (Bixler Run near Loysville), a residential stream (Spring Creek near Dauphin), and an orchard stream (Latimore Creek in Adams County) were 0.00, 0.03, 0.60, and 0.47 μg/liter, respectively (Table 4). Data from the two studies cannot be compared statistically because they represent only four areas in the Common-

TABLE 1. *Chlorinated hydrocarbon residues in Pennsylvania waters—1974-76*

WQN No.	Location [1]	Date Sampled	Sample Size, g	Stream Flow,[2] cfs	Flow Condition[3,4]	Chlorinated Hydrocarbon Residues, Corrected for Recovery, ng/kg (ppt)					
						Aroclor 1242	Aroclor 1254	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	ΣDDT
		STREAM SOURCES									
PCB S-1	Schuylkill R. Pottstown	9-23-74	3535	450		-[4]	-	-	-	-	-
	(Douglasville Br.)	6-5-75	3625[5]	900		-	-	-	-	-	-
PCB S-2	Delaware R. Easton	9-23-74	3525	2000		-	-	-	-	-	-
	(Northampton St. Br.)	6-5-75	3625[5]	3200		-	-	-	-	-	-
MDA-1	Delaware R. Phila.	10-17-74	3595		B	70	260	420	90	110	620
	(below metal bank)	9-24-75	1745		A	70	110	40	90	11	60
104	Delaware R. Hancock, NY	9-19-74	3585		B	-	-	-	-	-	-
	Bucking Twp. (Rt. 191 Br.)	5-8-75	3530		A	-	-	-	-	-	-
123	Lehigh R. Easton	9-23-74	3503	1100		-	-	-	-	tr[6]	tr
	(Third St. Br.)	6-5-75	3630[7]	1550		-	-	-	-	-	-
124	Lehigh R. Bethlehem	9-23-74	3525	800		-	-	-	-	-	-
	(Hill-to-Hill Br.)	6-5-75	3605[5]	1200		-	-	-	-	-	-
143	Schuylkill R. Phila.	10-1-74	3513		A	350	150	-	-	-	-
	(Strawberry Mansion Br.)	9-29-75	1790		A	-	-	-	-	-	-
202	Susquehanna R. Harrisbg.	9-12-74	3610		C	-	-	-	-	-	-
	(Walnut St. Br.)	5-2-75	3630		A	-	-	-	-	-	-
214	Juniata R. Newport	9-12-74	3624		C	-	-	-	-	-	-
	(Rt. 34 Br.)	4-25-75	3640		A	-	-	-	-	-	-
232	Susquehanna R. Holtwood	9-11-74	3600			-	-	-	-	-	-
	(Rt. 372 bridge)	5-2-75	3615		A	-	-	-	-	-	-
301	N. Br. Sesq. Danville	10-2-74	3585		B	-	-	-	-	-	-
	(Rt. 54 bridge)	12-1-75	1791			-	-	-	-	-	-
305	N. Br. Sesq. Towanda	9-19-74	3595		B	-	-	-	-	-	-
	(U.S. 6 bridge)	5-8-75	3583		A	-	-	-	-	-	-
401	W. Br. Sesq. Lewisburg	10-2-74	3370		B	-	-	-	-	-	-
	(Rt. 45 bridge)	12-1-75	1814			-	-	-	-	-	-
601	Lake Erie Mill Creek Twp.	9-10-74	3439			-	-	-	-	-	-
	(Erie Waterworks intake)	10-1-75	1795		B	-	-	-	-	-	-
701	Monongahela R. Rankin	9-23-74	3475		B	-	tr	-	-	-	-
		11-25-75	1775			-	-	-	-	-	-
801	Allegheny R. Kensington	10-4-74	3159		C	-	-	-	-	-	-
	(left & right sides)	11-21-75	1702		A	-	-	-	-	-	-
803	Allegheny R. Parker	9-24-74	3360	10,000	A	-	-	-	-	-	-
902	Ohio R. Sewickley	9-23-74	3540		B	-	-	-	-	-	-
	(Sewickley Br.)	11-25-75	1802		B	-	-	-	-	-	-
904	Beaver R. Rochester	10-1-74	3410		B	-	-	-	-	-	-
		11-13-75	1817		B	-	-	-	-	-	-
		OPEN WATER RESERVOIRS									
	Prince Gallitzen St. Park (breast of dam)	8-5-75	1755		B	-	-	-	-	-	-
	Quemahoning Res. (breast of dam)	10-1-75	1790			-	-	-	-	-	-
	Lake Wallenpawpack, Wolts Cove	10-7-75	1640			-	-	-	-	-	-
		COMMUNITY WATER SUPPLIES									
	Centre Township (Greene Co.)[8]	8-13-76	3830			-	-	-	-	-	-
	Lewistown (Mifflin Co.) (see below)	9-29-75	1715			460	-	-	-	-	-
	Lock Haven (Clinton Co.) (see below)	8-26-75	1783			tr	-	-	-	-	-
	Masontown (Fayette Co., gas station) (see below)	8-25-76	3872			20	-	-	-	-	-
	Point Marion (Fayette Co.)	9-3-75	1750			-	-	-	-	tr	tr
	Pottstown (Montgomery Co.)	7-29-76	3853			tr	-	-	-	-	-
	Waynesburg (Greene Co.) (see below)	9-3-75	1791			-	-	6	9	60	75

[1] WQN = Water Quality Network. See literature reference 9.
[2] Instantaneous measurement taken at time of sampling; cfs = cubic feet per second.
[3] A = above, B = normal, C = below.
[4] No detectable residue.
[5] Rain in morning.
[6] Trace; see Table 3.
[7] Rain in morning; stream had slight turbidity.
[8] South fork of Ten Mile Creek above McCourtney Run contained 0.7 ppt dieldrin.

TABLE 2. *Community water supplies which had no detectable PCB or DDT analog residues, Pennsylvania, 1974-76*

Allentown
Altoona
Ambridge (Beaver Co.)
Bedford
Belfast Twp. (Fulton Co.)
Bellefonte (Centre Co.)
Biglerville (Adams Co.)
Birdsboro (Berks Co.)
Bloomsburg (Columbia Co.)
Blossburg (Tioga Co.)
Blythe (Schuylkill Co.)
Bradford (McKean Co.)
Brockway (Jefferson Co.)
Brookhaven (Delaware Co.)
Butler City
Cambridge Springs (Crawford Co.)
Canonsburg (Washington Co.)
Carmichaels Boro (Greene Co., 1976; plant influent, plant effluent at Amoco, distrib. system at Amoco on S. Vine and at 106 S. Vine)
Center Township (Greene Co., 1976; McCourtney Run, Lightner Run, Pursley Cr., Rush Run)
Chambersburg (Franklin Co.)
Charleroi (Washington Co.)
Columbia (Lancaster Co.)
Conemaugh (Somerset Co.)
Cumberland Twp. (Greene Co., 1976)
Danville (Montour Co.)
Delaware Water Gap (Monroe Co.)
Derry (Westmoreland Co.)
Derry Twp. (Dauphin Co., 1975 and 1976, 1883 g)
Duncannon (Perry Co.)
Dunkard (Greene Co., 1976; Dunkard Cr., Brunley Res., Gogallatto Res., plant influent, plant effluent, Monongahela R. up from Cheat R.)
Eaglesmere (Sullivan Co.)
East Berlin (Adams Co.)
Ellwood City (Lawrence Co.)
Emlenton (Venango Co.)
Emporium (Cameron Co.)
Erie
Fayette City (Fayette Co.)
Franklin Twp. (Greene Co., 1976; Brown's Cr., first and second unnamed tribs. to Ten Mile Cr., Clear Run)
Gallitzin (Cambria Co.)
German Twp. (Fayette Co., 1976; Cat's Cr., Big Run filtration plant, plant effluent, distrib. system at Kolenlick's Sunoco at the Pizza Parlor)
Gettysburg (Adams Co.)
Greensboro (Greene Co., 1976)
Greenville (Mercer Co.)
Hallstead (Sesquehanna Co.)
Harrisburg
Hazelton
Hershey
Hollidaysburg (Blair Co.)
Homer City (Indiana Co.)
Honesdale (Wayne Co.)
Houtzdale (Clearfield Co.)
Jersey Shore (Lycoming Co., 1975 and 1976)
Johnstown
Kingston (Lucerne Co.)
Lancaster
Lansford (Carbon Co.)
Lebanon
Lewistown (Mifflin Co., 1976; Laurel Run, Mutterbaugh Gap Run, distrib. system at Burnham M. A. STP)
Lititz (Lancaster Co.)
Littlestown (Adams Co.)
Lock Haven (1976; Castanea Res., Keller Res., Rosecranes Res., distrib. system at Swinehart residence in McElhatten and at R and K Grocery)
Lower Paxton Twp. (Dauphin Co., 1976)
Masontown Boro (Fayette Co., 1976; plant influent, plant effluent, distrib. system at 305 S. Main St.)
McKeesport
Media (Delaware Co.)
Middleburg (Synder Co.)
Mifflinburg (Union Co.)
Milesburg (Centre Co.)
Milford (Pike Co.)
Milton (Northumberland Co.)
Monongahela Twp. (Greene Co., 1976)
Morrisville (Bucks Co.)
Moscow (Lackawana Co.)
Mount Joy (Lancaster Co.)
New Bethelem (Clarion Co.)
New Freedom (York Co.)
Nicholson (Wyoming Co.)
Nicholson Twp. (Fayette Co., 1976)
North East (Erie Co.)
North Shenango (Crawford Co.)
Northampton (Northampton Co.)
Oakmont (Allegheny Co., 1976)
Parker City (Armstrong Co.)
Philadelphia (Belmont Res., Queen Lane Res., Torresdale Res., Phila. suburban at Lower Merion pump station)
Pittsburgh
Point Marion (Fayette Co., 1976; first unnamed trib. to Cheat R., Grassy Run, Conrad Res., Rose Res., plant influent, plant effluent)
Rauchtown (Clinton Co., 1976)
Reading
Ridgway (Elk Co.)
Roulette (Potter Co.)
Saltsburg (Indiana Co., 1976)
Scranton
Sharon (Mercer Co., 1976)
Shawnee State Park (Bedford Co.)
Sheffield (Warren Co.)
Shrewsbury (York Co.)
Springhill Twp. (Fayette Co., 1976)
Stewartstown (York Co.)
Stroudsburg (Monroe Co.)
Tionesta (Forest Co.)
Towanda (Bradford Co.)
Union Twp. (Mifflin Co., 1976)
Wagontown (Chester Co.)
Waynesburg (Greene Co., 1976; plant influent, plant effluent, distrib. system at Gulf Station and at McDonald's Rest.)
West Penn Power (Greene Co., 1976; intake, Monongehela R. below discharge)
Wilkes-Barre
Williamsport
Woolrich (Clinton Co., 1976)
Wormsleyburg (Cumberland Co.)
York

NOTE: Samples collected August-November 1975 were all approx. 1800 grams. Samples collected July-August 1976 are marked "1976" and were all approx. 3600 grams, unless designated otherwise.

TABLE 3. *Detectability limits of method of analysis for water samples*

Limit	Sample Size, g	Aroclor 1242	Aroclor 1254	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	∑DDT
No detectable residue	3600	<10	<10	<1	<1	<1	—
	1800	<20	<20	<2	<2	<2	—
Trace	3600	10–25	10–25	1–2.5	1–2.5	1–2.5	—
	1800	20–50	20–50	2–5	2–5	2–5	—

TABLE 4. *Comparison of mean ΣDDT in Pennsylvania waters, 1969–76*

Study Area	Residues, μg/liter	
	N	Means
FROM TRUHLAR AND REED, 1969–71[1]		
Forest stream	10	0.00
Farm stream	14	0.03
Residential stream	24	0.60
Orchard stream	32	0.47
FROM THIS WORK, 1975–76	Residues, μg/kg	
Rivers	19[2]	0.018
Communities	110	0.0007

[1] See literature reference 22.
[2] Sampled twice.

wealth. One can say only that DDT concentration in water in forest and farm areas is similar to that in rivers across Pennsylvania: 0.018 μg/kg. DDT concentration in water in the forest area is similar to that in areas serving the communities of Pennsylvania: 0.0007 μg/kg.

DETERMINING STREAM POLLUTION

Direct point source sampling of water in streams gives the analyst only limited information regarding the presence of contaminants over a broad geographic area such as a state. Unless a large number of samples are taken at a high cost, other sampling means must be used.

Several averaging methods are available. These include continuous or intermittent direct sampling at one location, or indirect sampling. The content of lypophobic compounds, e.g., chlorinated hydrocarbons such as PCBs, in water can be estimated by analyzing the fish in that water or by analyzing the organic sediment in the active stream system. Since both media are organic, the equilibria of these compounds in water are in the direction of the former.

Truhlar and Reed analyzed bottom sediment separately for DDT analogs (*22*). They found a positive correlation of ΣDDT between the bottom sediment and the water of Lattimore Creek.

Another time-averaged method would be the analysis of fish, an animal high on the biomagnification chain. Selecting the fish based on its feeding habits or range of habitat enables the researcher to evaluate the concentration of PCBs or DDT analogs present over the fish's lifetime.

The Pennsylvania Department of Environment Resources (DER) conducted a survey of the levels of PCBs and other chlorinated hydrocarbons in fish from selected Pennsylvania waters (*5*) in 1976. Many of the collection points correspond closely to those for water-stream sources in the Pennsylvania Water Quality Network. Pertinent results of that study are summarized in Table 5.

In the DER study, fish were generally sampled 6 or 18 months after water sampling times. The range in PCB concentrations (ΣPCB expressed as Aroclor 1254) for sunfish (carnivorous feeders living on zooplankton and aquatic invertebrates) was 0.05–0.86 μg/g (wet weight, whole-tissue). PCBs were found in nine stations corresponding to WQN sampling points. ΣDDT and analog concentration in sunfish ranged from 0.01 to 0.10 μg/g and were found in only seven of these stations.

The range in PCB concentrations for carp, which are bottom feeders, was greater: 0.04–1.63 μg/g in 12 stations. For DDT the range was also greater: 0.01–1.27 μg/g in nine stations.

An examination of the points of correspondence for the fish and water studies showed little correlation of data. Correlation coefficients (Pearson product moment) for water versus sunfish for PCBs and DDT were undefined and −0.156, respectively. Those for water versus carp for PCBs and DDT were 0.132 and 0.891, respectively. Such poor correlations are probably due to the fact that most water samples contained less than a determinable amount of contaminant. The correlation of DDT in water versus carp (0.891) is misleading, even though the hypothesis of no correlation is rejected at the 1 percent level since 12 of the 16 values lie essentially at the origin of the plot.

MODEL OF PCB FLOW IN STREAMS

Several models could be designed to determine how PCB's, DDT, polybrominated biphenyls, and similar pollutants are transported in waterways. In one such model, compounds are present in more or less constant concentrations. This could result from a slow dissolution of a concentrated source into the water flowing by the source. The absolute level could vary from one point to another, but would remain at a given level for several days or weeks or would change slowly from level to level.

Another model that could describe this information is a slug-flow model. In this type, the compounds are released within a matter of hours as a result of some human or natural activity. This contamination moves downstream at a rate close to the water movement. At any point along the stream the concentration of contaminants could vary widely from moment to moment.

The slug-flow model would have to be modified to account for the effects of sediment usually present in stream flows. PCBs moving downstream adsorb to organic matter up to the capacity of the matter. Given only a small amount of lyphophobic contaminant and enough sediment and stream turbulence, the entire amount could be adsorbed by the organic material. The rate of transfer from water to sediment would depend on the concentration of organic matter and turbulence. The rate would probably be relatively high due to the minimal solubility in water. Hence if small amounts of PCB enter a flowing stream, soon they could be ad-

TABLE 5. *PCB and ΣDDT residues in fish taken from locations corresponding to water sampling locations, Pennsylvania, April–July, 1976* [1]

Fish Sampling Location	WQN Station at Water Sampling Location[2]	Residues, μg/g Wet Weight, Whole Fish					
		Sunfish[3]			Carp[4]		
		No. Fish	PCB	ΣDDT	No. Fish	PCB	ΣDDT
I–10	PCB S-1	10	0.56	0.02	2	1.63	ND [5]
I–19	PCB S-2	5	ND	ND	5	ND	ND
I–1	MDA-1				4	0.70	1.27
I–14	123	6	ND	ND	7	0.61	ND
I–15	124	13	ND	ND	5	ND	ND
I–5	143				5	0.61	0.18
III–1	202	5	0.86	0.08	5	ND	ND
III–2	232	5	0.06	0.01	5	0.07	0.01
III–4	301	5	0.72	ND	5	0.66	0.04
III–9	305	5	0.05	0.01	5	1.16	0.07
III–10	401	6	0.07	0.02	5	ND	ND
IV–7	601	5	0.19	ND	5	0.93	0.33
IV–21 W	701				5	ND	0.36
IV–21 E					5	0.63	0.73
IV–8	801	5	0.10	0.10	5	0.31	0.01
IV–9	803	5	ND	ND	5	0.04	ND
IV–4	902	5	0.81	0.03	5	0.76	0.02
IV–4	904	5	0.81	0.03	5	0.76	0.02

[1] See literature reference 5.
[2] WQN = Water Quality Network. See literature reference 9.
[3] Includes smallmouth, largemouth, and rock bass; black and white crappie; bluegill; and pumpkinseed and redbreast sunfish.
[4] Includes goldfish, channel catfish, brown bullhead, white sucker, and silver redhorse.
[5] ND = no detectable residue, <0.01 μg/g.

sorbed to sediment. If the sediment settles, the PCB would not be found in the analysis of the sample taken downstream from that point.

The present study included 19 major stream stations sampled twice, 7 to 14 months apart. From this collection of data, there were seven examples of residues in the first sampling (PCBs or DDT analogs) and not in the second. Only one station had residues (Station MDA-1, Delaware River, Philadelphia) in both samplings. These data, although limited, support slug-flow movement.

The Truhlar and Reed (*22*) data support this conclusion more strongly. There were numerous occasions when the authors analyzed two to four samples taken the same day. Considering only those that contained significant amounts of DDT analogs, there were five examples that could support a slug model of transport. In these cases the levels found varied widely. Some were also inversely proportional to sediment concentration; the Truhlar and Reed data as well as the data reported here included sediment concentrations in the water samples. Only three examples supported a constant content model, i.e., the levels found were relatively constant or they varied proportionally with sediment concentration.

The sampling procedures of this study and the Truhlar and Reed study were different. Truhlar and Reed picked a few sites and took many samples for two years. In this study, many sites across the State were sampled, but none more than twice. Both studies appear to support a slug-flow model for contamination.

Both models could be operating at the same time. Slugs of contamination could be imposed on a continuous level of the material. The concentration at the continuous level may be lower than has been routinely detected; if so, greater analytical sensitivity is needed.

In this study, most samples were 1.8 kg which is a convenient size to collect and ship. The detection limit for this size was 20 ppt (ng/kg) for PCBs and 2 ppt for each DDT analog. These compounds are insoluble in water at levels slightly above two powers of ten higher for Aroclor 1254 and three to four powers of ten higher for Aroclor 1242 (*11*). In a recent ruling to improve sensitivity in water analyses, the U.S. Environmental Protection Agency set the ambient water criterion (*25*) for PCBs in navigable waters at 1 ng/liter. This would require a detection limit of at least 1/10 as much or 0.1 ng/liter, which is 1/200 of the limit obtained in this work. To lower this detection limit, the sample size would need to be increased, the background levels would need to be decreased, and the detection technique would have to be improved.

By examining the bioaccumulation of PCBs in fish, one can gain insight in this matter. One could assume that the PCB level in water was constant at the upper end of the nondetectable level, i.e., 10 ppt in this work. At a bioaccumulation factor of 274,000 for fathead minnows (*25*), fish would contain 2.7 ppm PCBs. Nine stations in the Pennsylvania study yielded carp containing 0.13–1.63 ppm (*5*), and no stations yielded carp containing greater than 1.63 ppm. Since no samples of fish contained as much as 2.7 ppm, it is reasonable that no water samples contained even detectable amounts. This was, in fact, the case. Excluding input by sediment or other means this would indicate that the minimum detectable levels in water were too

high by a factor of ten to produce these concentrations in fish. Hence the detectability limit must be decreased from 10 to 0.1. When this is done, the question of whether the PCB levels are constant or whether they appear in slugs as suggested in this paper can be approached.

A single sample analysis of stream water would be appropriate if the source of contamination were continuous. If the source were variable, however, either multigrab samples or a time-averaged method would be required to determine the contamination profile.

Acknowledgments

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GENERAL

DDT Residues in Rainwater in New Brunswick and Estimate of Aerial Transport of DDT into the Gulf of St. Lawrence, 1967-68

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ABSTRACT

Residues of DDT were detected in 76 of 101 samples of rainwater collected during spring and summer at several sites in the Province of New Brunswick in 1967 and 1968, and at one site in the Magdalen Islands, Quebec, in 1968. The dominant residue was p,p'-DDT. Levels of DDT and metabolites combined ranged from < 0.01 to 1.33 μg/kg. Levels of DDT and metabolites in the pollen of four species of forest trees in New Brunswick ranged from 0.544 to 1.01 mg/kg; such contaminated pollen possibly contributed to residues in rainwater. Residue data for rainwater from two sites were used to estimate the amount of DDT aerially transported into the Gulf of St. Lawrence during July to October 1968.

Introduction

Despite the interest in transport of organochlorines through the atmospheric environment, few residue data collected in areas remote from pesticide use have been related to specific spray programs. Levels of DDT and its metabolites in rainwater sampled at a number of sites in New Brunswick in 1967 and 1968, and at one station in Quebec in 1968, are reported. Also recorded are concentrations of DDT in the pollen of four species of trees common in the New Brunswick forest, since contaminated pollen could have contributed to residues in rainwater samples. Investigations were conducted at a time when there was still a substantial regional use of DDT in forestry and agriculture. By determining residue levels in some rainwater samples, one may estimate the amount of DDT transported through the atmosphere into the adjacent Gulf of St. Lawrence.

Materials and Methods

SAMPLE COLLECTION AND ANALYTICAL PROCEDURE

Fifty-eight rainwater samples were collected at seven sites in New Brunswick from mid-May to late July 1967. The sites were selected to encircle the area of forest in the center of the Province to be sprayed with DDT and the organophosphates phosphamidon and fenitrothion. In 1968, 43 samples were collected from mid-April to mid-August from four sites in New Brunswick and one in the Magdalen Islands, Quebec, all downwind from the treatment area. New Brunswick is about 90 percent forested. Most collection sites in that Province were in or near the forest. The Magdalen Islands are located in the middle of the Gulf of St. Lawrence, about 250 km from the eastern shore of New Brunswick. The geographic positions of the collection sites are given in Tables 1 and 2.

Water was collected in glass bottles through a large glass funnel supported in a section of stove pipe firmly placed in the ground. In 1967, authors attempted to keep the sample size to about 1 liter and used both clear and amber bottles as well as a small fiber-glass pad to prevent possible clogging of the funnel stem by leaves and large insects. The following year, authors modified the procedure slightly to exclude the fiber-glass pad, since there had been no obvious accumulation of extraneous organic matter in bottles or funnel stems. In 1968, amber bottles were used at all stations except one, which was supplied with clear bottles wrapped in aluminum foil. Precautions were taken whenever possible to prevent contamination of the samples. All glassware was washed in detergent, and rinsed several times with water and twice with redistilled acetone before use.

In May 1968, before the forest had been sprayed, authors obtained samples of pollen of jack pine (*Pinus banksiana*), red spruce (*Picea rubens*), white birch (*Betula papyrifera*), and red maple (*Acer rubrum*) from a region in central New

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TABLE 1. *DDT residue levels in rainwater samples from New Brunswick, Canada—1967*

Site	Collection Period	Total Volume, ml	Residues, μg/kg			
			p,p'-DDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Allardville 47 28 65 29	15-19 May	1150	ND	ND	ND	ND
	19-27 May	2000	ND	ND	ND	ND
	27-28 May	1180	0.09	0.06	0.09	0.14
	28 May-8 June	1280	0.10	0.08	0.06	0.14
	8-22 June	1220	0.03	0.08	ND	0.09
	22-26 June	1910	0.06	0.05	ND	0.12
	26 June-4 July	1590	0.02	<0.01	ND	<0.01
	4-10 July	1180	ND	ND	ND	ND
	10-13 July	154	0.19	0.21	0.14	0.36
St. Louis 46 44 64 58	15-18 May	1140	ND	ND	ND	ND
	18-19 May	1400	ND	ND	ND	ND
	19-26 May	1070	ND	ND	ND	ND
	26-29 May	1110	ND	ND	ND	ND
	29 May-15 June	1070	0.03	0.06	ND	0.14
	15-19 June	1150	0.03	0.05	0.04	0.09
	19-24 June	1100	0.37	0.18	2.78[1]	20.0[1]
	24 June-1 July	1170	0.19	0.24	0.72[1]	12.5[1]
	1-5 July	1060	ND	0.06	ND	0.81[1]
	5-8 July	1040	ND	<0.01	0.07	0.53[1]
	8-13 July	1380	0.06	0.07	ND	0.78[1]
Canaan 46 05 65 22	12-13 May	1850	ND	ND	ND	ND
	13-18 May	1570	ND	ND	ND	ND
	18-26 May	1380	ND	ND	ND	ND
	26-29 May	1230	<0.01	ND	ND	ND
	29 May-12 June	1120	ND	ND	ND	ND
	12-18 June	2176	ND	ND	ND	ND
	18-25 June	1560	0.04	0.05	0.02	0.06
	25 June-3 July	1210	ND	ND	ND	ND
	3-4 July	1600	0.06	0.06	ND	0.08
	4-14 July	710	ND	ND	ND	ND
Fredericton 45 58 66 39	12-18 May	1140	0.06	0.06	0.07	0.17
	18-26 May	1340	ND	0.05	0.02	0.08
	26 May-17 June	1370	0.04	0.06	ND	ND
	17-19 June	1130	0.04	0.05	0.02	0.09
	19-24 June	1640	0.03	0.03	ND	0.10
	24 June-4 July	1010	0.21	0.17	0.09	0.41
	4-26 July	980	0.03	0.05	ND	0.08
Parker Ridge 46 27 66 31	13-20 May	1520	<0.01	<0.01	ND	<0.01
	20-27 May	1200	ND	<0.01	ND	<0.01
	27 May-12 June	1210	ND	ND	ND	0.27
	12-18 June	1360	0.03	0.06	0.05	0.31
	18-25 June	1130	ND	ND	ND	0.50
	25-26 June	1040	ND	ND	ND	ND
	26 June-2 July	1240	ND	ND	ND	ND
	2-9 July	1400	ND	ND	ND	ND
	9-24 July	1000	ND	ND	ND	ND
Gordonsville 46 29 67 31	16-20 May	970	ND	ND	ND	ND
	20 May-8 June	1020	ND	ND	ND	ND
	8-17 June	1180	ND	ND	ND	ND
	17-24 June	1100	0.03	0.03	0.01	0.08
	24-26 June	1930	ND	ND	ND	ND
	26 June-10 July	1940	0.06	ND	ND	ND
Kedgwick 47 39 67 21	16-20 May	1210	0.04	0.06	0.04	0.09
	20 May-2 June	1030	<0.01	0.03	ND	0.09
	2-23 June	970	0.02	ND	ND	0.17
	23-26 June	1930	ND	ND	ND	ND
	26 June-14 July	1780	0.04	0.06	ND	0.29
	14-18 July	425	0.06	0.12	0.06	0.22

NOTE: ND = not detected = <0.001 μg/kg.
[1]Local contamination.

Brunswick which had been aerially sprayed with DDT each year from 1956 to 1967, except 1966. The collection procedure was simply to shake staminate flowers carefully in thoroughly cleaned glass jars.

Pesticide residues were determined by gas-liquid chromatography (GLC). The organochlorine compounds were detected with an electron-capture (EC) detector; the EC detector was used in conjuction with the phosphate (P) detector for the organophosphates. A 500-ml aliquot of each sample was extracted with ether-hexane in the presence of sodium sulfate, the phases were allowed to separate, and the aqueous portion was re-extracted with hexane. The combined organic phases were rinsed with distilled water, dried over anhydrous sodium sulfate, and analyzed.

TABLE 2. *DDT residue levels in rainwater samples from New Brunswick and Quebec, Canada—1968*

Site	Collection Period	Total Volume, ml	Residues, μg/kg			
			p,p'-DDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Miscou	7-27 May	690	0.01	ND	ND	0.01
47 01	27 May-4 June	1050	ND	ND	ND	0.02
64 31	4-19 June	1000	<0.01	ND	ND	0.01
	19-24 June	1110	0.08	ND	ND	0.01
	24 June-7 July	1640	0.13	ND	ND	0.04
	7-24 July	960	<0.01	ND	0.01	0.11
	24 July-9 August	700	<0.01	ND	<0.01	0.08
Escuminac	10-29 May	380	ND	ND	0.42	0.91
47 04	29 May-1 June	615	ND	ND	0.04	0.08
64 48	1-5 June	640	ND	ND	ND	0.03
	5-16 June	1015	<0.01	ND	0.02	0.06
	16-20 June	860	0.01	ND	0.04	0.06
	20-22 June	1260	0.01	ND	0.03	0.01
	22 June-1 July	1550	ND	ND	ND	0.01
	1-14 July	430	ND	ND	ND	ND
	14-20 July	730	ND	ND	ND	0.05
	20 July-9 August	380	ND	ND	ND	0.05
Clairville	19-29 April	1925	<0.01	0.01	ND	0.01
46 23	29 April-9 May	400	0.02	ND	ND	0.02
65 06	9-22 May	960	0.03	<0.01	ND	0.01
	22 May-11 June	1700	0.03	<0.01	ND	0.01
	11-12 June	530	0.02	<0.01	ND	0.01
	12-17 June	760	0.01	0.01	ND	0.01
	17-20	760	0.01	0.01	ND	0.01
	20-23 June	1590	0.01	ND	ND	0.01
	23 June-1 July	1675	ND	ND	ND	ND
	1-26 July	555	0.31	0.21	0.60[1]	10.4[1]
Fredericton	17-23 April	580	0.01	ND	ND	0.06
45 58	23-28 April	950	<0.01	ND	ND	0.06
66 39	28 April-14 May	480	0.01	ND	ND	0.06
	14-22 May	1015	0.01	0.02	0.02	0.13
	22 May-3 June	715	0.01	0.03	0.03	0.16
	3-5 June	620	<0.01	0.02	0.02	0.09
	5-17 June	487	0.01	0.02	0.01	0.11
	17-22 June	1700	0.01	0.01	0.01	0.08
	22 June-6 July	1900	0.01	0.01	ND	0.02
	6 July-11 August	1960	0.01	0.01	ND	0.07
Magdalen	18 May-2 June	1120	ND	ND	ND	0.01
Islands	2-15 June	980	0.01	<0.01	ND	0.03
47 23	15-16 June	970	<0.01	ND	ND	<0.01
61 52	16-17 June	1130	<0.01	<0.01	ND	<0.01
	17-23 June	990	0.02	<0.01	ND	0.01
	23 June-13 August	1925	ND	ND	ND	0.06

NOTE: ND = not detected = <0.001 μg/kg.
[1] Local contamination.

Reagent grade chemicals were used. Ether was redistilled but hexane (Nanograde) was used as supplied. Varian Aerograph Models 1200 (equipped with an EC detector) and 204 (with EC and P detectors) were used. The column effluent from the Model 204 was split 50:50 so that half the injected sample passed through each detector, and the responses were recorded by an attached dual-pen recorder. Both glass helix GLC columns, 152 cm × 0.32 cm, were packed with 4 percent SE-30/60 percent QF-1 on 60-80 mesh acid-washed Chromosorb W.

An investigation was made to ascertain what portion, if any, of the pesticides adhered to the bottle surface and therefore was not quantitatively recovered. The water sample was removed from each of four bottles and the bottle surfaces were thoroughly rinsed with 200 ml of acetone. The acetone extracts were analyzed. The results prompted a modification of the method in order to recover the residues remaining on the bottle surfaces. Each bottle was thereafter emptied and rinsed with 200 ml acetone, and an appropriate proportion of the rinsings was added to the 500-ml aliquot to be analyzed. All residues reported here include those adsorbed on bottle surfaces. No attempt was made to determine what residues remained on the funnel surfaces. A jar cleaned in the same manner as those used for collecting the pollen samples was also rinsed with acetone, and no additional residues were recovered.

The recoveries of added pesticides including lindane, heptachlor, aldrin, heptachlor epoxide, DDE, dieldrin, TDE, *p,p'*-DDT, and fenitrothion were essentially 100 percent, and although the confidence level for the method is taken as 0.01 ppb (μg/kg), residues as low as 0.001 ppb were detected. Confirmatory analyses were conducted with thin-layer chromatography (TLC). A small peak with a retention time similar to fenitrothion was observed with the

EC detector in some samples, but the P detector proved the absence of any organophosphate. During the analyses, authors were aware of the possibility of interferences from polychlorinated biphenyls (PCBs). The GLC peaks eluting after DDT were small, indicating that if PCBs (Aroclor 1254 and 1260 types) were present, the amounts were small. The TLC analyses confirmed that the organochlorine pesticides were responsible for at least the major portion of the residues.

TABLE 3. *Precipitation in the Magdalen Islands, Canada—1968*

MONTH	PRECIPITATION, CM
January	11.0
February	6.6
March	7.6
April	6.1
May	3.5
June	9.4
July	3.4
August	12.0
September	2.8
October	8.1
November	11.4
December	12.4

CALCULATION OF DDT DEPOSITION INTO THE GULF OF ST. LAWRENCE

The concentrations of ΣDDT in the rainwater at the two downwind sites most remote from the spray program and from possible local usage, viz., Miscou and the Magdalen Islands, were used to estimate deposition of DDT into the Gulf of St. Lawrence. Monthly precipitation in the Magdalen Islands in 1968 is shown in Table 3. The initial assumption made was that the falloff of concentration with the distance from source was exponential, and the geometry of the region was reduced to a wedge-shaped area (Fig. 1).

Thus the concentration is considered to be related to the distance from the area of forest spraying by the equation $y = Ae^{-bx}$, where y = concentration of DDT, x = distance, and A and b are constants. Each segment of the wedge has an area of Kx.dx, where $K = 2\pi a/360$ and a = the angle of the wedge. L is the approximate distance between central New Brunswick and the eastern coastline of that province, and U is the distance between central New Brunswick and the arc of a circle running through southwestern Newfoundland. The amount of DDT deposited per centimeter of rainfall in the truncated wedge-shaped area extending from the coastline of New Brunswick to southwestern Newfoundland is:

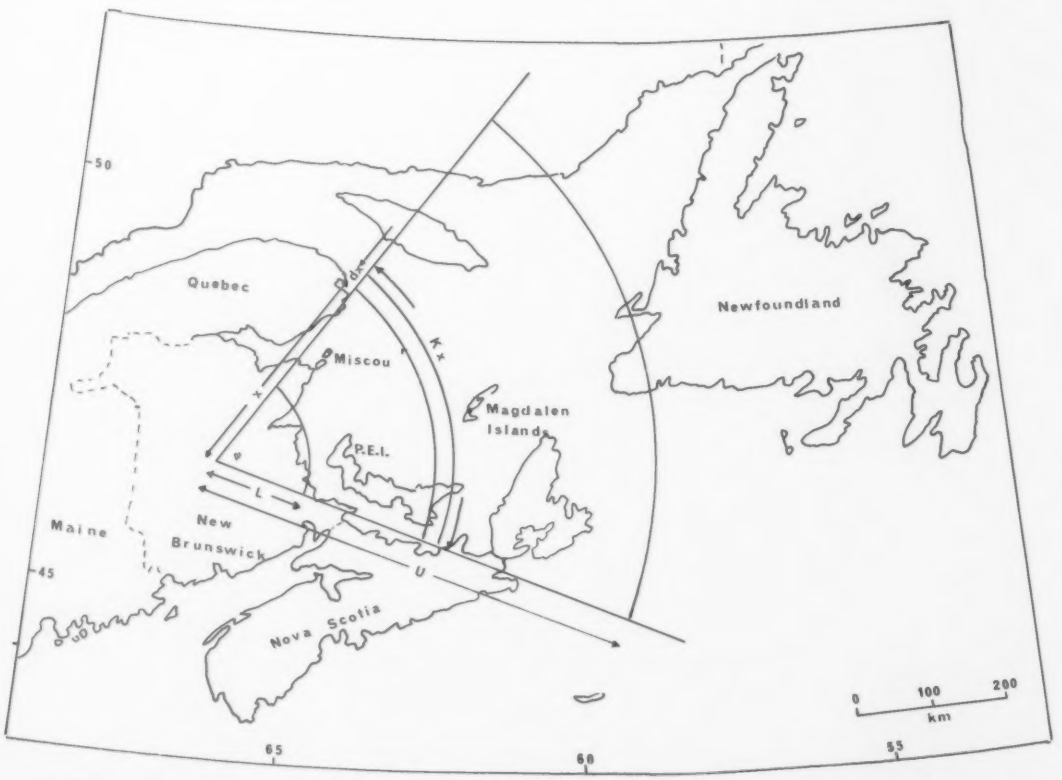

FIGURE 1. *Map of the Atlantic Provinces showing method of estimating DDT deposition into the Gulf of St. Lawrence*

$$\int_L^U f(x) . Kx.dx =$$

$$\int_L^U Ae^{-bx} Kx.dx = KA \int_L^U xe^{-bx} dx$$

Integrating by parts gives:

$$\text{Amount of DDT} = KA/b^2(Lb + 1)e^{-Lb} - (Ub + 1)e^{-Ub}$$

Results and Discussion

The 1967 and 1968 analysis results for the DDT group residues in rainwater are shown in Tables 1 and 2, respectively. DDT and/or its metabolites were detected in 76 of the 101 samples analyzed. In 1967, ΣDDT ranged from <0.01 to 0.90 μg/kg. In 1968, ΣDDT ranged from <0.01 to 1.33 μg/kg.

The dominant residue in almost all rainwater samples was *p,p'*-DDT. Using the Friedman two-way analysis of variance, *p,p'*-DDT has a rank sum of 344.5, compared to 245 for DDE, 228 for TDE, and 192.5 for *o,p'*-DDT. The ranking difference for *p,p*-DDT was significant at $P < 0.0001$ over all other compounds. Similiar rankings were found at individual sites; *p,p'*-DDT was the dominant residue in 11 of 12 groups. The differences in the ranking between the remaining residues were not significant. The percentage of *o,p'*-DDT was variable, but in half the groups, the percentage was between 10 and 20. The relatively small proportion of DDE shows that little DDT has been incorporated in biological tissue to be subsequently released to the atmosphere. The occurence of TDE may be due to photodecomposition of DDT *(8)*. The ratios of the various compounds of the DDT group are similar to those reported from Great Britain in the mid-1960s (*1, 10*). The levels reported here range higher than do other published values for that period (Table 4).

The only other organochlorine compound detected was dieldrin; small concentrations (0.01 μg/kg) were found in five rainwater samples from two sites in 1967 and in 11 samples from four sites in 1968. Airborne dieldrin could have originated as dieldrin or from epoxidation of aldrin. The only potential source in the region was the agricultural use of aldrin in New Brunswick and adjacent areas for the control of root maggots (*Hylemya* spp.) in cruciferous crops.

Central New Brunswick was sprayed June 10-29, 1967, and June 4-28, 1968, to protect the forest against damage by spruce budworm (*Choristoneura fumiferana*). The aircraft operated 15 – 45 m above the tree canopy. Approximate ΣDDT and organophosphate emissions in thousands of kg active ingredient were, respectively, 190 and 57 in 1967 and 20 and 52 in 1968. About 40 thousand kg DDT was used in northern Maine in 1967 for spruce budworm control. None was used for that purpose in 1968. In western and northwestern New Brunswick and also in neighboring Prince Edward Island and Aroostook County, Maine, potato-growing is an important industry. Among the many pests of that crop, the flea beetle (*Epitrix cucumeris*) and, to a lesser extent, the Colorado potato beetle (*Leptinotarsa decemlineata*) were, at the time of the study, controlled by ground application of DDT in late June and early July. About 27 thousand kg DDT was used annually in New Brunswick for that purpose.

The prevailing wind during the collection periods was from the west and southwest. Meteorological data taken at the collection sites did not indicate a relation between peak residue concentrations in rainwater samples and periods of maximum insecticide use. The periods in June during which the samples were collected were likely out of phase

TABLE 4. *Comparison of DDT and metabolite levels found in precipitation in other investigations, 1965-74*

Region	Year	Residues, μg/kg			Reference
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	
Great Britain	1965	0.002-0.017	0.016-0.034	0.113-0.195	Abbott et al. *(1)*
U.S.A. (Ohio)	1965	0.005-0.030	not given	0.070-0.340	Cohen and Pinkerton *(3)*
Great Britain	1966-1967	0.007-0.028	0.007-0.034	0.018-0.066	Tarrant and Tatton *(10)*
Antarctica	1966-1967	ND	ND	0.040	Peterle *(7)*
Eastern Canada	1967-1968	ND-0.21[1]	ND-0.21[1]	ND-0.91[1]	This study
U.S.A. (Hawaii)	1970-1971	ND	not given	0.001-0.13	Bevenue et al. *(2)*
U.S.A. (New York)	1974	ND-trace	ND	ND-0.002	Peakall *(6)*

NOTE: ND = not detected.
[1] Excludes known local contamination.

with those during which forest spray aircraft were active, since no spraying took place when it was raining. It is possible, therefore, that insecticide drifting downwind from spray zones was not intercepted by rain at the collection sites. The occurrence of measurable amounts of DDT in rainwater before the spray programs started suggests more distant sources of contamination and also suggests that sprayed forest and agricultural areas in the region may contaminate the atmosphere for many months after spray programs are completed. Spencer et al. *(9)* concluded that a high percentage of organochlorine pesticides lost from plant surfaces is lost by volatilization. Lloyd-Jones *(5)* examined the rate of evaporation of DDT from planchets and screens and concluded that about half the DDT applied to field crops may enter the atmosphere. He found that the rate of evaporation was highly temperature-dependent. Thus the amount of DDT lost from surfaces during the cold New Brunswick winter months presumably would be very small. The dispersion of tree pollen over considerable distances has been documented by several investigators and reviewed by Lanner *(4)*. Concentrations of DDT and metabolites ranged from 0.544 to 1.01 mg/kg in tree pollen from the forest of central New Brunswick in 1968 (Table 5). Pollen was taken when some of the earliest, pre-spray samples of rainwater were collected that year. If present, such contaminated pollen may have contributed residues to the levels reported for those rainwater samples, since pollen was not separated from water before extraction and analysis.

It seems likely that DDT used in forestry and agriculture in the Maritime Provinces and Maine contributed to fallout of that pesticide in rain in the region. However, for the reasons outlined and since there was no clear spatial or temporal pattern of residue peaks in the rainwater samples, the contamination cannot be related directly to regional use of DDT during the period of investigation.

By using the equations developed above and mean values of DDT concentration in rain of 0.11 μg/kg at Miscou and 0.06 μg/kg at the Magdalen Islands, it can be calculated that about 5,000 kg DDT were deposited in the wedge-shaped area covering the Gulf of St. Lawrence from July to October in 1968. The time frame was chosen on the assumption that the evaporation of DDT from ground and plant surfaces would be severely reduced after the onset of cold weather in November. The calculation of DDT deposition is a rough one only, and completely depends on a very small number of measurements. The assumption that 0.06 μg/kg is typical of DDT fallout at the Magdalen Islands during that period cannot be tested. Although the calculations presented are only first approximations, they do support the current view that the aerial route is important in the transport of organochlorines in the environment.

TABLE 5. *DDT residue levels in tree pollen from central New Brunswick, Canada—1968*

Species	Residues, mg/kg			
	p,p'-DDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Jack pine	0.025	0.047	0.050	0.887
Red spruce	0.109	0.083	0.117	0.565
White birch	0.108	0.056	0.110	0.270
Red maple	0.116	0.077	0.107	0.504

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We are much indebted to J. A. Keith, Canadian Wildlife Service, Department of Fisheries and the Environment, who conceived the project and was involved in technical discussions during this study. G.E.J. Smith, also of that agency, provided valuable advice on analysis and application of the residue data. Information on precipitation was supplied by the Atmospheric Environment Service, Department of Fisheries and the Environment.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

AROCLOR 1242	PCB, approximately 42% chlorine
AROCLOR 1254	PCB, approximately 54% chlorine
CHLORDANE	1,2,3,5,6,7,8,8-Octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene. The technical product is a mixture of several compounds including heptachlor, chlordene, and two isomeric forms of chlordane.
DDE	Dichlorodiphenyl dichloro-ethylene (degradation product of DDT) *p, p'*-DDE: 1,1-Dichloro-2,2-bis(*p*-chlorophenyl) ethylene *o,p'*-DDE: 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethylene
DDT	Main component (*p,p'*-DDT): α-Bis(*p*-chlorophenyl) β,β,β-trichloroethane. Other isomers are possible and some are present in the commercial product. *o,p'*-DDT: [1,1,1-Trichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7:8,8a-octahydro-1,4-*endo-exo*-5,8-dimethanonaphthalene
ENDRIN	Hexachloroepoxyoctahydro-*endo,endo*-dimethanonaphthalene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro 2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
PCBs (Polychlorinated Biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
TDE	2,2-Bis(*p*-chlorophenyl)-1,1-dichloroethane (including isomers and dehydrochlorination products)

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SUBJECT AND AUTHOR INDEXES

Volume 11, June 1977—March 1978

Preface

Primary headings in the subject index include pesticide compounds, media in which pesticide residues are monitored, and major concepts related to the monitoring of pesticides in the environment. Pesticide compounds are listed by common names; trade names are used for those which have no common names.

Secondary headings cross-reference the primary headings. For a paper which discusses five or more organochlorines the compounds are grouped by class under media and concept headings but each compound appears individually under the primary headings for pesticide compounds.

In the author index all information on a paper appears in the senior author's citations: associate authors, title of the paper, and volume, issue, and pages where the article was published. Names of associate authors are cross-referenced as minor headings, but the reader is referred to the senior author's entry for the paper's complete citation.

SUBJECT INDEX

A

Aldrin (see also Dieldrin)
- Degradation
  - 11(2)88-93
  - 11(2):94-98
- Diet, Total
  - 11(3):116-131
- Factors Influencing Residues
  - 11(2):88-93
  - 11(2):94-98
  - 11(3):138-145
  - 11(4):199-204
- Food and Feed
  - 11(2):88-93
  - 11(2):94-98
  - 11(4):161-164
- Plants (other than those used for food and feed)
  - 11(2):99-106
  - 11(4):199-204
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):88-93
  - 11(2):94-98
  - 11(2):99-106
- Water
  - 11(2):99-106
  - 11(4):199-204
- Wildlife
  - 11(2):99-106
  - 11(3):138-145

Aroclor (see also PCB's)
- Wildlife
  - 11(1):40-53
  - 11(2):69-87

Aroclor 1242 (see also PCB's)
- Water
  - 11(4):190-198

Aroclor 1254 (see also PCB's)
- Water
  - 11(4):190-198
- Wildlife
  - 11(3):134-137

Aroclor 1260 (see also PCB's)
- Wildlife
  - 11(1):40-53

Arsenic
- Diet, Total
  - 11(3):116-131
- Sediment
  - 11(4):182-189
- Wildlife
  - 11(1):5-34
  - 11(1):35-39
  - 11(1):40-53
  - 11(4):182-189

Azodrin
- Plants (other than those used for food and feed)
  - 11(2):99-106
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):99-106
- Water
  - 11(2):99-106
- Wildlife
  - 11(2):99-106

B

BHC/Lindane
- Degradation
  - 11(2):88-93
- Diet, Total
  - 11(3):116-131
- Factors Influencing Residues
  - 11(2):88-93
- Food and Feed
  - 11(2):88-93
  - 11(4):161-164
- Humans
  - 11(1):54-55
- Plants (other than those used for food and feed)
  - 11(2):99-106
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):88-93
  - 11(2):99-106
- Water
  - 11(2):99-106
- Wildlife
  - 11(2):99-106

Botran
- Diet, Total
  - 11(3):116-131

C

Cadmium
- Diet, Total
  - 11(3):116-131
- Sediment
  - 11(4):182-189
- Wildlife
  - 11(1):5-34
  - 11(1):35-39
  - 11(1):40-53
  - 11(4):182-189

Carbaryl
- Diet, Total
  - 11(3):116-131
- Plants (other than those used for food and feed)
  - 11(2):99-106
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):99-106
- Water
  - 11(2):99-106
- Wildlife
  - 11(2):99-106

Carbofuran
- Plants (other than those used for food and feed)
  - 11(2):99-106
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):99-106
- Water
  - 11(2):99-106
- Wildlife
  - 11(2):99-106

Chlordane
- Diet, Total
  - 11(3):116-131
- Food and Feed
  - 11(4):161-164
- Plants (other than those used for food and feed)
  - 11(2):99-106
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):99-106
- Water
  - 11(2):99-106
- Wildlife
  - 11(1):40-53
  - 11(2):99-106
  - 11(3):134-137

Chloroform
- Wildlife
  - 11(3):134-137

Chromium
- Wildlife
  - 11(1):40-53

CIPC
- Diet, Total
  - 11(3):116-131

Copper
- Wildlife
  - 11(1):40-53

D

DCPA
- Diet, Total
  - 11(3):116-131

DDD, see TDE

DDE
- Diet, Total
  - 11(3):116-131
- Factors Influencing Residues
  - 11(1):40-53
  - 11(2):61-63
  - 11(3):111-115
  - 11(3):138-145
  - 11(4):170-181
  - 11(4):199-204
- Food and Feed
  - 11(4):161-164
- Humans
  - 11(1):1-4
  - 11(1):54-55
  - 11(2):61-63
  - 11(3):111-115
- Plants (other than those used for food and feed)
  - 11(2):99-106
  - 11(4):199-204
- Sediment
  - 11(2):99-106
- Soil
  - 11(2):99-106
- Water
  - 11(2):99-106
  - 11(4):190-198
  - 11(4):199-204
- Wildlife
  - 11(1):40-53
  - 11(2):99-106
  - 11(3):134-137
  - 11(3):138-145
  - 11(4):170-181

DDT
Diet, Total
11(3):116-131
Factors Influencing Residues
11(1):40-53
11(2):61-63
11(3):111-115
11(3):138-145
11(4):165-169
11(4):199-204
Food and Feed
11(4):161-164
Humans
11(1):1-4
11(1):45-55
11(2):61-63
11(3):111-115
Plants (other than those used for food and feed)
11(2):99-106
11(4):199-204
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
11(4):190-198
11(4):199-204
Wildlife
11(1):40-53
11(2):99-106
11(3):134-137
11(3):138-145
11(4):165-169

Degradation
Aldrin
11(2):88-93
11(2):94-98
BHC/Lindane
11(2):88-93
Dieldrin
11(2):88-93
11(2):94-98
Endrin
11(2):88-93
Mirex
11(3):146-156

Diazinon
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Dicofol
Diet, Total
11(3):116-131

Dieldrin (see also Aldrin)
Degradation
11(2):88-93
11(2):94-98
Diet, Total
11(3):116-131
Factors Influencing Residues
11(1):40-53
11(2):88-93
11(2):94-98
11(3):111-115
11(3):138-145
11(4):199-204
Food and Feed
11(2):88-93
11(2):94-98
11(4):161-164
Humans
11(3):111-115
Plants (other than those used for food and feed)
11(2):99-106
11(4):199-204
Sediment
11(2):99-106
Soil
11(2):88-93
11(2):94-98
11(2):99-106
Water
11(2):99-106
11(4):199-204
Wildlife
11(1):40-53
11(2):99-106
11(3):134-137
11(3):138-145

E

Endosulfan
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Endrin
Degradation
11(2):88-93
Diet, Total
11(3):116-131
Factors Influencing Residues
11(2):88-93
11(4):170-181
Food and Feed
11(2):88-93
11(4):161-164
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):88-93
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106
11(4):170-181

Ethion
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Ethyl Parathion
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

F

Factors Influencing Residues
Age
DDE
11(2):61-63
11(3):111-115
11(4):170-181
DDT
11(2):61-63
11(3):111-115
11(4):165-169
dieldrin
11(3):111-115
endrin
11(4):170-181
PCB's
11(4):170-181
Environmental, Geographical and Locational
aldrin
11(3):138-145
11(4):199-204
DDE
11(3):111-115
11(3):138-145
11(4):170-181
11(4):199-204
DDT
11(3):111-115
11(3):138-145
11(4):199-204
dieldrin
11(3):111-115
11(3):138-145
11(4):199-204
endrin
11(4):170-181
heptachlor
11(3):138-145
heptachlor epoxide
11(3):138-145
PCB's
11(4):170-181
TDE
11(4):199-204
Farming Practices and Land Use
aldrin
11(2):88-93
11(2):94-98
11(3):138-145
11(4):199-204
BHC/lindane
11(2):88-93
DDE
11(3):138-145
11(4):199-204
DDT
11(3):138-145
11(4):199-204
dieldrin
11(2):88-93
11(2):94-98
11(3):138-145
11(4):199-204
endrin
11(2):88-93

heptachlor
11(3):138-145
heptachlor epoxide
11(3):138-145
TDE
11(4):199-204
Race
DDE
11(2):61-63
DDT
11(2):61-63
Seasonal and Temporal
aldrin
11(3):138-145
11(4):199-204
DDE
11(3):138-145
11(4):170-181
11(4):199-204
DDT
11(3):138-145
11(4):199-204
dieldrin
11(3):138-145
11(4):199-204
endrin
11(4):170-181
heptachlor
11(3):138-145
heptachlor epoxide
11(3):138-145
PCB's
11(4):170-181
TDE
11(4):199-204
Sediment
aldrin
11(3):138-145
DDE
11(3):138-145
DDT
11(3):138-145
dieldrin
11(3):138-145
heptachlor
11(3):138-145
heptachlor epoxide
11(3):138-145
Sex
DDE
11(3):111-115
11(4):170-181
DDT
11(3):111-115
dieldrin
11(3):111-115
endrin
11(4):170-181
PCB's
11(4):170-181
Species
aldrin
11(3):138-145
DDE
11(3):138-145
DDT
11(3):138-145
dieldrin
11(3):138-145
heptachlor
11(3):138-145
heptachlor epoxide
11(3):138-145
Weight and Size
DDE
11(4):170-181
DDT
11(4):165-169
endrin
11(4):170-181
PCB's
11(4):170-181

Food and Feed

Beverages
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Dairy Products
aldrin
11(4):161-164
BHC/lindane
11(4):161-164
chlordane
11(4):161-164
DDE
11(4):161-164
DDT
11(4):161-164
dieldrin
11(4):161-164
endrin
11(4):161-164
heptachlor
11(4):161-164
heptachlor epoxide
11(4):161-164
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
TDE
11(4):161-164
Diet, Total
aldrin
11(3):116-131
arsenic
11(3):116-131
BHC/lindane
11(3):116-131
botran
11(3):116-131
cadmium
11(3):116-131
carbaryl
11(3):116-131
chlordane
11(3):116-131
CIPC
11(3):116-131
DCPA
11(3):116-131
DDE
11(3):116-131
DDT
11(3):116-131
diazinon
11(3):116-131
dicofol
11(3):116-131
dieldrin
11(3):116-131
endosulfan
11(3):116-131
endrin
11(3):116-131
ethion
11(3):116-131
HCB
11(3):116-131
heptachlor epoxide
11(3):116-131
lead
11(3):116-131
leptophos
11(3):116-131
lindane
11(3):116-131
malathion
11(3):116-131
mercury
11(3):116-131
methoxychlor
11(3):116-131
methyl parathion
11(3):116-131
nonachlor
11(3):116-131
octachlor epoxide
11(3):116-131
orthophenylphenol
11(3):116-131
parathion
11(3):116-131
PCA
11(3):116-131
PCB
11(3):116-131
PCNB
11(3):116-131
PCP
11(3):116-131
pentachlorobenzene
11(3):116-131
perthane
11(3):116-131
phosalone
11(3):116-131
ronnel
11(3):116-131
selenium
11(3):116-131
TCNB
11(3):116-131
TDE
11(3):116-131
toxaphene
11(3):116-131
zinc
11(3):116-131
Fruits
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Grain and Cereal Products
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Meat, Fish, and Poultry
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Oils, Fats, and Shortening
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Soybeans
aldrin
11(2):94-98
dieldrin
11(2):94-98

Sugars and Adjuncts
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131
Vegetables
aldrin
11(2):88-93
11(2):94-98
BHC/lindane
11(2):88-93
dieldrin
11(2):88-93
11(2):94-98
endrin
11(2):88-93
metals
11(3):116-131
organochlorines
11(3):116-131
organophosphates
11(3):116-131

G

Guthion
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

H

HCB
Diet, Total
11(3):116-131
Wildlife
11(1):40-53

Heptachlor
Factors Influencing Residues
11(3):138-145
Food and Feed
11(4):161-164
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106
11(3):138-145

Heptachlor Epoxide
Diet, Total
11(3):116-131
Factors Influencing Residues
11(3):138-145
Food and Feed
11(4):161-164
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106
11(3):134-137
11(3):138-145

Hexachlorobenzene
Wildlife
11(3):134-137

Humans
Blood
BHC/lindane
11(1):54-55
DDE
11(1):54-55
11(3):111-115
DDT
11(1):54-55
11(3):111-115
dieldrin
11(3):111-115
TDE
11(1):54-55
Children
DDE
11(1):1-4
DDT
11(1):1-4
Cholesterol
DDE
11(3):111-115
DDT
11(3):111-115
dieldrin
11(3):111-115
Creatine
DDE
11(3):111-115
DDT
11(3):111-115
dieldrin
11(3):111-115
Glucose
DDE
11(3):111-115
DDT
11(3):111-115
dieldrin
11(3):111-115
Tissues
DDE
11(2):61-63
DDT
11(2):61-63
Uric Acid
DDE
11(3):111-115
DDT
11(3):111-115
dieldrin
11(3):111-115

L

Lead
Diet, Total
11(3):116-131
Sediment
11(4):182-189
Wildlife
11(1):5-34
11(1):35-39
11(1):40-53
11(4):182-189

Leptophos
Diet, Total
11(3):116-131

Lindane (see BHC)

M

Magnesium
Wildlife
11(1):40-53

Malathion
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Mercury
Diet, Total
11(3):116-131
Sediment
11(4):182-189
Wildlife
11(1):5-34
11(1):35-39
11(1):40-53
11(3):132-133
11(4):182-189

Methomyl
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Methoxychlor
Diet, Total
11(3):116-131

Methyl Parathion
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Methyl Trithion
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

Mirex
Degradation
11(3):146-156
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(1):40-53
11(2):64-68
11(2):99-106
11(3):134-137
11(3):146-156

N

Nickel
Wildlife
11(1):40-53

Nonachlor
Diet, Total
11(3):116-131
Wildlife
11(1):40-53
11(3):134-137

O

Octachlor Epoxide
Diet, Total
11(3):116-131

Orthophenylphenol
Diet, Total
11(3):116-131

Oxychlordane
Wildlife
11(3):134-137

P

Parathion
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

PCA
Diet, Total
11(3):116-131

PCB's
Diet, Total
11(3):116-131
Factors Influencing Residues
11(4):170-181
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(1):40-53
11(2):69-87
11(2):99-106
11(3):134-137
11(4):170-181

PCNB
Diet, Total
11(3):116-131

PCP
Diet, Total
11(3):116-131

Pentachlorobenzene
Diet, Total
11(3):116-131

Perthane
Diet, Total
11(3):116-131

Phosalone
Diet, Total
11(3):116-131

Plants (other than those used for food and feed)
Tobacco
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Trees and Shrubs
aldrin
11(4):199-204
DDE
11(4):199-204
DDT
11(4):199-204
dieldrin
11(4):199-204
TDE
11(4):199-204

R

Ronnel
Diet, Total
11(3):116-131

S

Sediment
Lakes and Ponds
aldrin
11(2):99-106
arsenic
11(4):182-189
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
cadmium
11(4):182-189
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
lead
11(4):182-189
malathion
11(2):99-106
mercury
11(4):182-189

methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Reservoirs
arsenic
11(4):182-189
cadmium
11(4):182-189
lead
11(4):182-189
mercury
11(4):182-189

Selenium
Diet, Total
11(3):116-131
Wildlife
11(1):5-34
11(1):35-39
11(1):40-53

Soil
Croplands
aldrin
11(2):88-93
11(2):94-98
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):88-93
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):88-93
11(2):94-98
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):88-93
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106

Strobane
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(2):99-106

T

TCNB
Diet, Total
11(3):116-131

TDE
Diet, Total
11(3):116-131
Factors Influencing Residues
11(1):40-53
11(4):199-204
Food and Feed
11(4):161-164
Humans
11(1):54-55
Plants (other than those used for food and feed)
11(2):99-106
11(4):199-204
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
11(4):190-198
11(4):199-204
Wildlife
11(1):40-53
11(2):99-106
11(3):134-137

Toxaphene
Diet, Total
11(3):116-131
Plants (other than those used for food and feed)
11(2):99-106
Sediment
11(2):99-106
Soil
11(2):99-106
Water
11(2):99-106
Wildlife
11(1):40-53
11(2):99-106
11(3):134-137

W

Water (see also Sediment)
Drinking
aroclor 1242
11(4):190-198
aroclor 1254
11(4):190-198
DDE
11(4):190-198
DDT
11(4):190-198
TDE
11(4):190-198
Lakes and Ponds
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106

TDE
11(2):99-106
toxaphene
11(2):99-106
Rain
aldrin
11(4):199-204
DDE
11(4):199-204
DDT
11(4):199-204
dieldrin
11(4):199-204
TDE
11(4):199-204
Rivers and Streams
aroclor 1242
11(4):190-198
aroclor 1254
11(4):190-198
DDE
11(4):190-198
DDT
11(4):190-198
TDE
11(4):190-198

Wildlife
Amphibians
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
11(3):146-156
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Aquatic
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Birds
aroclor 1254
11(3):134-137
arsenic
11(1):35-39
11(1):40-53
cadmium
11(1):35-39
11(1):40-53
chlordane
11(3):134-137
chloroform
11(3):134-137
chronium
11(1):40-53
copper
11(1):40-53
DDE
11(1):40-53
11(3):134-137
11(4):170-181
DDT
11(1):40-53
11(3):134-137
dieldrin
11(1):40-53
11(3):134-137
endrin
11(4):170-181
heptachlor epoxide
11(1):40-53
11(3):134-137
hexachlorobenzene
11(3):134-137
lead
11(1):35-39
11(1):40-53
magnesium
11(1):40-53
mercury
11(1):35-39
11(1):40-53
metals
11(1):35-39
11(1):40-53
mirex
11(1):40-53
11(2):64-68
11(3):134-137
11(3):146-156
nickel
11(1):40-53
nonachlor
11(1):40-53
11(3):134-137
organochlorines
11(1):40-53
oxychlordane
11(3):134-137
PCB's
11(1):40-53
11(3):134-137
11(4):170-181
selenium
11(1):35-39
11(1):40-53
TDE
11(1):40-53
11(3):134-137
toxaphene
11(1):40-53
11(3):134-137
zinc
11(1):40-53
Clams
arsenic
11(4):182-189
cadmium
11(4):182-189
lead
11(4):182-189
mercury
11(4):182-189
Fish
aldrin
11(2):99-106
11(3):138-145
aroclor
11(2):69-87
arsenic
11(1):5-34
azodrin
11(2):99-106

BHC/lindane
11(2):99-106
cadmium
11(1):5-34
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
11(3):138-145
DDT
11(2):99-106
11(3):138-145
11(4):165-169
diazinon
11(2):99-106
dieldrin
11(2):99-106
11(3):138-145
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
11(3):138-145
heptachlor epoxide
11(2):99-106
11(3):138-145
lead
11(1):5-34
malathion
11(2):99-106
mercury
11(1):5-34
11(3):132-133
metals
11(1):5-34
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
11(3):146-156
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):69-87
11(2):99-106
selenium
11(1):5-34
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Insects
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
11(3):146-156
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Invertebrates
mirex
11(3):146-156
Mammals
mirex
11(3):146-156
Plankton
arsenic
11(4):182-189
cadmium
11(4):182-189
lead
11(4):182-189
mercury
11(4):182-189
Reptiles
aldrin
11(2):99-106
azodrin
11(2):99-106
BHC/lindane
11(2):99-106
carbaryl
11(2):99-106
carbofuran
11(2):99-106
chlordane
11(2):99-106
DDE
11(2):99-106
DDT
11(2):99-106
diazinon
11(2):99-106
dieldrin
11(2):99-106
endosulfan
11(2):99-106
endrin
11(2):99-106
ethion
11(2):99-106
ethyl parathion
11(2):99-106
guthion
11(2):99-106
heptachlor
11(2):99-106
heptachlor epoxide
11(2):99-106
malathion
11(2):99-106
methomyl
11(2):99-106
methyl parathion
11(2):99-106
methyl trithion
11(2):99-106
mirex
11(2):99-106
11(3):146-156
organochlorines
11(2):99-106
organophosphates
11(2):99-106
parathion
11(2):99-106
PCB's
11(2):99-106
strobane
11(2):99-106
TDE
11(2):99-106
toxaphene
11(2):99-106
Shellfish
arsenic
11(4):182-189
cadmium
11(4):182-189
lead
11(4):182-189
mercury
11(4):182-189

Z

Zinc
Diet, Total
11(3):116-131
Wildlife
11(1):40-53

AUTHOR INDEX

A

Amore, F. J., see Wedberg, J. L.

B

Banks, W. A., see Wheeler, W. B.
Bean, Jerry R., see Walsh, David F.
Bean, Jerry R., see White, Donald H.
Belisle, Andre A., see Prouty, Richard M.
Bender, Thomas R., see Serat, William F.
Berger, Bernard L., see Walsh, David F.
Bloomer, Arthur W., Nash, Stanley I., Price, Harold A., and Welch, Robert L. A study of pesticide residues in Michigan's general population, 1968-70. 11(3):111-115
Blus, Lawrence J., Neely, Burkett S., Jr., Lamont, Thair G., and Mulhern, Bernard. Residues of organochlorines and heavy metals in tissues and eggs of brown pelicans, 1969-73. 11(1):40-53
Bond, C. A., see Reeves, R. G.
Brown, Ralph, see Wheeler, W. B.
Bruce, W. N., see Moore, Steve, III
Bulkley, Ross Vivian. Variations in DDT concentration in muscle tissue of channel catfish, *Ictalurus punctatus*, from the Des Moines River, 1971. 11(4):165-169
Burks, John M., see Serat, William F.

C

Cromartie, Eugene, see Prouty, Richard M.

E

Ellarson, Robert S., see Peterson, Steven R.

F

Ferguson, James, see Serat, William F.

G

Ganyard, M. C., see Reeves, R. G
Gerhart, Ellen Heath. Concentrations of total mercury in several fishes from Delaware Bay. 11(3):132-133

H

Hair, J. D., see Kendall, Ronald J.
Hamelink, Jerry Lee, see Vanderford, Michael James

J

Jackson, H. B., see Kendall, Ronald J.
Johnson, R. D., and Manske, D. D. Pesticide and other chemical residues in Total Diet samples (XI). 11(3):116-131
Jouvenaz, D. P., see Wheeler, W. B.

K

Kaiser, T. Earl, see Prouty, Richard M.
Kendall, Ronald J., Noblet, R., Hair, J. D., and Jackson, H. B. Mirex residues in bobwhite quail after aerial application of bait for fire ant control, South Carolina—1975-76. 11(2):64-68
Knight, Luther A., Jr., see Price, Richard E.
Krauthacker, Blanka, see Reiner, Elsa
Kuhlman, D. E., see Moore, Steve, III
Kurtz, David A. Residues of PCB's and DDT in Pennsylvania streams, community water supplies, and reservoirs, 1974–76. 11(4):190–198
Kutz, F. W., Yobs, A. R., Strassman, S. C., and Viar, J. F., Jr. Effects of reducing DDT usage on total DDT storage in humans. 11(2):61–63

L

Lamont, Thair G., see Blus, Lawrence J.
Lamont, Thair G., see Prouty, Richard M.
Lee, Min K., see Serat, William F.
Locke, Louis N., see Prouty, Richard M.
Lofgren, C. S., see Wheeler, W. B.
Longcore, Jerry R., see White, Donald H.

M

McAvoy, Harold, see Wedberg, J. L.
Manske, D. D., see Johnson, R. D.
Mengle, Donald C., see Serat, William F.
Moore, Steve, III, Petty, H. B., Bruce, W. N., Randell, Roscoe, and Kuhlman, D. E. Dieldrin residues in soybeans in Illinois, 1965, 1966, 1967, 1971, and 1974. 11(2):94–98
Moore, Steve, III, see Wedberg, J. L.
Mulhern, Bernard, see Blus, Lawrence J.
Mulhern, Bernard M., see Prouty, Richard M.

N

Nash, Stanley I., see Bloomer, Arthur W.
Neely, Burkett S., Jr., see Blus, Lawrence J.
Nesbitt, Steve, see Wheeler, W. B.
Noblet, R., see Kendall, Ronald J.

P

Peakall, D. B., see Pearce, P. A.
Pearce, P. A., Reynolds, L. M., and Peakall, D. B. DDT residues in rainwater in New Brunswick and estimate of aerial transport of DDT into the Gulf of St. Lawrence. 11(4):199–205
Peterson, Steven R., and Ellarson, Robert S. *p, p'*-DDE, PCB's, and endrin in oldsquaw ducks, 1969–73. 11(4):170–181
Petty, H. B., see Moore, Steve, III.
Price, Richard E., and Knight, Luther A., Jr. Mercury, cadmium, lead, and arsenic in sediments, plankton, and clams from Lake Washington and Sardis Reservoir, Mississippi. 11(4):182–189
Price, Harold A., see Bloomer, Arthur W.
Prouty, Richard M., Reichel, William L., Locke, Louis N., Belisle, Andre A., Cromartie, Eugene, Kaiser, T. Earl, Lamont, Thair G., Mulhern, Bernard M., and Swineford, Douglas M. Residues of organochlorine pesticides and polychlorinated biphenyls and autopsy data for bald eagles, 1973–74. 11(3):134–137

R

Randell, Roscoe, see Moore, Steve, III
Reeves, R. G., Woodham, D. W., Ganyard, M. C., and Bond, C. A. Preliminary monitoring of agricultural pesticides in a cooperative tobacco pest management project in North Carolina, 1971—first-year study. 11(2):99–106
Reichel, William L., see Prouty, Richard M.
Reiner, Elsa, Krauthacker, Blanka, Stipcevic, Marinko, and Stefanac, Zlata. Blood levels of chlorinated hydrocarbon residues in the population of a continental town in Croatia (Yugoslavia). 11(1):54–55
Reynolds, L. M., see Pearce, P.A.

S

Serat, William F., Lee, Min K., Van Loon, Albert J., Mengle, Donald C., Ferguson, James, Burks, John M., and Bender, Thomas R. DDT and DDE in the blood and diet of Eskimo children from Hooper Bay, Alaska. 11(1):1–4
Skinner, Lawrence, C., see Spagnoli, John J.

Spagnoli, John J., and Skinner, Lawrence C. PCB's in fish from selected waters of New York State. 11(2):69–87
Stefanac, Zlata, see Reiner, Elsa
Stipcevic, Marinko, see Reiner, Elsa
Strassman, S. C., see Kutz, F. W.
Suzuki, M., Yamato, Y., and Watanabe, T. Organochlorine insecticide residues in field soils of the Kitakyushu District, Japan—1970–74. 11(2):88–93
Swineford, Douglas M., see Prouty, Richard M.

V

Vanderford, Michael James, and Hamelink, Jerry Lee. Influence of environmental factors on pesticide levels in sport fish. 11(3):138–145
Van Loon, Albert J., see Serat, William F.
VanMiddelem, C. H., see Wheeler, W. B.
Viar, J. F., Jr., see Kutz, F. W.

W

Walsh, David F., Berger, Bernard L., and Bean, Jerry R. Mercury, arsenic, lead, cadmium, and selenium residues in fish, 1971–73—National Pesticide Monitoring Program. 11(1):5–34
Watanabe, T., see Suzuki, M.
Wedberg, J. L., Moore, S., III, Amore, F. J., and McAvoy, Harold. Organochlorine insecticide residues in bovine milk and manufactured milk products in Illinois, 1971–76. 11(4):161-164
Welch, Robert L., see Bloomer, Arthur W.
Wheeler, W. B., Jouvenaz, D. P., Wojcik, Daniel P., Banks, W. A., VanMiddelem, C. H., Lofgren, C. S., Nesbitt, Steve, Williams, Lovett, and Brown, Ralph. Mirex residues in nontarget organisms after application of 10–5 bait for fire ant control, northeast Florida—1972–74. 11(3):146-156.
White, Donald H., Bean, Jerry R., and Longcore, Jerry R. Nationwide residues of mercury, lead, cadmium, arsenic, and selenium in starlings, 1973. 11(1):35–39
Williams, Lovett, see Wheeler, W. B.
Wojcik, Daniel P., see Wheeler, W. B.
Woodham, D. W., see Reeves, R. G.

Y

Yamato, Y., see Suzuki, M.
Yobs, A. R., see Kutz, F. W.

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